Comparison of mathematical models of the kinetic of drying pennyroyal leaves

*(Mentha pulegium L.)*

Comparação de modelos matemáticos da cinética da secagem de folhas de pennyroyal (*Mentha pulegium L.*)

Comparación de modelos matemáticos de cinética de secado de hojas de poleo (*Mentha pulegium L.*)

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**Abstract**

*Mentha pulegium L.*, popularly known as pennyroyal, has simple leaves that give off a pleasant aroma when crushed. The main objective of this work was to carry out the drying of pennyroyal leaves, to estimate the effective diffusion coefficient through drying kinetics in forced convection, and to determine the best mathematical model at four different temperatures (40, 50, and 60 °C) inflow. 1.5 m/s air. Analyzing the drying curves, it was observed that the drying kinetics were strongly influenced by temperature. The thin layer models that best fit the experimental data were Approximate Diffusion, Two Terms, and Logarithmic for the temperatures of 40, 50, and 60 °C, respectively. The evaluation method used the R² (coefficient of determination), RMSE (root-mean-square), and X² (chi-square), and the coefficient of determination parameter remained >0.90. The effective diffusion coefficient decreased 74% with increasing temperature from 40 °C to 60 °C and enthalpy and entropy decreased with increasing temperature, while Gibb's free energy increased 5% for each increment of 10 °C in temperature.

**Keywords:** Convection heat transfer; Mathematical modeling; Drying curves.
Resumo

A *Mentha pulegium* L. popularmente conhecida como poejo, possui folhas simples que expelem um aroma agradável quando são trituradas. Este trabalho teve como principal objetivo realizar a secagem das folhas de poejo, estimar por meio de cinética de secagem em convecção forçada, o coeficiente de difusão efetivo e determinar o melhor modelo matemático em quatro diferentes temperaturas (40, 50 e 60 °C) em fluxo de ar de 1,5 m/s. Analisando as curvas de secagem observou-se que a cinética de secagem foi fortemente influenciada pela temperatura. Os modelos de camada fina que melhor se ajustaram aos dados experimentais foram Difusão Aproximada, Dois Termos e Logarítmico para as temperaturas de 40, 50 e 60 °C, respectivamente. O método de avaliação deu-se utilizando o R² (coeficiente de determinação), RMSE (raiz quadrada do erro médio) e Χ² (Qui-quadrado), sendo que o parâmetro de coeficiente de determinação se manteve >0,90. O coeficiente de difusão efetivo diminuiu 74% com a elevação da temperatura de 40 °C para 60 °C e a entalpia e a entropia decresceram com o aumento da temperatura, enquanto a energia livre de Gibbs aumentou 5% para cada incremento de 10 °C na temperatura.

Palavras-chave: Secagem por convecção; Modelo matemático; Curvas de secagem.

Resumen

*Mentha pulegium* L. conocida popularmente como poleo, tiene hojas simples que al triturarlas desprenden un agradable aroma. El objetivo principal de este trabajo fue realizar el secado de hojas de poleo, estimar, mediante cinéticas de secado por convección forzada, el coeficiente de difusión efectivo y determinar el mejor modelo matemático a cuatro temperaturas diferentes (40, 50 y 60 °C) en flujo de agua, aire a 1,5 m/s. Al analizar las curvas de secado, se observó que la cinética de secado estaba fuertemente influenciada por la temperatura. Los modelos de capa fina que mejor se ajustaron a los datos experimentales fueron Difusión Aproximada, Dos Términos y Logarítmico para temperaturas de 40, 50 y 60 °C, respectivamente. El método de evaluación se realizó mediante R² (coeficiente de determinación), RMSE (raíz cuadrada del error medio) y Χ² (Chi-cuadrado), y el parámetro del coeficiente de determinación se mantuvo >0,90. El coeficiente de difusión efectivo disminuyó en un 74 % al aumentar la temperatura de 40 °C a 60 °C y la entalpía y la entropía disminuyeron al aumentar la temperatura, mientras que la energía libre de Gibbs aumentó en un 5 % por cada incremento de 10 °C en la temperatura.

Palabras clave: Secado por convección; Modelo matemático; Curvas de secado.

1. Introduction

The drying of food products can be deliberated as a simultaneous procedure of heat and mass transfer between the product and the drying air, which consists in the removal of excessive moisture contained within the material through dissipation, caused by forced air convection heated, to allow for greater conservation of quality during storage for long seasons (Chua et al., 2000; Da Silva Morais et al., 2013; Tavone et al., 2021).

The use of mathematical models in the drying process helps researchers to better optimize, integrate and control energy during the drying process. When the water activity (Aw) is reduced to the minimum level, where a balance and stability point of free water within the food is found, chemical and biological deterioration tend to be minimized, a process that helps in food preservation and storage for long periods (Mghazli et al., 2017).

*Mentha pulegium* L. known in some places in Brazil as pennyroyal is a plant used as a drug in several countries due to its medicinal properties. Its composition is rich in bioactive compounds, such as antioxidants and phenolic compounds, anti-inflammatory, analgesic, digestive action (Ahmed et al., 2018; Mollaei et al., 2020; Yakoubi et al., 2021). The use of dried plant extracts turns out to be more viable, as it increases shelf life and facilitates the storage of this plant material.

Therefore, the aim of this study was (1) to evaluate the effect of drying by convection at different temperatures, (2) to verify which is the best mathematical model that fits in drying, (3) to calculate the effective diffusion coefficient and thermodynamic properties for drying pennyroyal leaves.

2. Methodology

Plant Material

*Mentha pulegium* L. (pennyroyal) was acquired through the living laboratory of Alternative Agriculture practices at Faculdade Intercultura Indígena - FAIND, located at the Federal University of Grande Dourados (Mato Grosso do Sul – Brazil). The pennyroyal was selected according to the color of the leaves (preferably green), which eliminated those with physical
damage, then they were washed and sanitized in 1% sodium hypochlorite for 15 min. After this time, the penny royal leaves were stored under refrigeration at a temperature of 5 ºC in polyethylene bags.

Drying

The drying process was carried out with a tray dryer, at temperatures of 40, 50, and 60 ºC and a speed of 1.5 m/s, until obtaining a constant temperature, which varied for each temperature studied. Before drying, the initial moisture of the penny royal leaves was determined through the method of drying in an oven until reaching constant weight (AOAC, 1990). The dryer used is at the laboratory level, consisting of a drying chamber where the trays (NG Scientific brand) are placed, and the samples are deposited in these. The air pre-heating system is provided by a set of electrical resistances and an air circulation system consisting of a fan. An anemometer is used to control the speed of hot air that was inserted inside the dryer.

Before drying the samples, the dryer was turned on half an hour beforehand to stabilize the temperature. Once the temperature was stabilized, the trays containing the penny royal leaves were placed inside the dryer compartment, to start the drying process. The samples were removed from the dryer during the first hour at 15-minutes intervals, then at 1-hour intervals until constant dynamic equilibrium in the Ubu samples (moisture on a wet basis) was lower than 10%, drying was carried out in triplicate for every temperature.

Moisture content and mathematical models

The different moisture contents according to the time interval and their weighing was calculated from the difference between the initial weight and the weighing, considering the weight loss. The moisture contents at different temperatures were converted about moisture (MR), according to equation 1. It is noteworthy that the MR is dimensionless.

\[
MR = \frac{(M_x - M_{x0})}{(M_{xi} - M_{x0})} \quad (eq. 1)
\]

where MR is the water content ratio (dimensionless value), Mx is the water content of the product represented on a dry basis (bs); Mx 0 is the equilibrium water content of the product (bs), and Mxi is the initial water content of the product (bs).

Statistical Parameters

The mathematical models used in the analysis of the drying kinetics were selected according to the literature, and the approximate diffusion, Two Terms, Logarithmic, Henderson & Pabis, Newton, and Page models were chosen, as shown in Table 1.

| Model Name          | Model Designation        | Equations   |
|---------------------|--------------------------|-------------|
| Approximate Diffusion| MR=a exp(-kt)+(1-a) exp(-kbt) | (eq. 2)     |
| Two Terms           | MR = a exp(-k1t) + b exp(-k2t) | (eq. 3)     |
| logarithmic         | MR=a exp(-kt)+c          | (eq. 4)     |
| Henderson & Pabis   | MR = the exp(-kt)        | (eq. 5)     |
| Newton              | MR=exp(-kt)              | (eq. 6)     |
| Page                | MR=exp(-ktn)             | (eq. 7)     |

Source: Authors.

Diffusion coefficient (Def) and influence of temperature on Def

The diffusion coefficient was calculated using Equation 7, based on the theory of liquid diffusion, and the Arrhenius equation was used to evaluate the influence of temperature on the effective diffusion coefficient.
Where: \( D_i = \) effective diffusion coefficient (m\(^2\) s\(^{-1}\)); \( L = \) product thickness (m); \( t = \) drying time (s); \( n = \) number of terms in the model. \( D_0 = \) pre-exponential factor; \( E_a = \) activation energy (kJ mol\(^{-1}\)); \( R = \) universal gas constant (8.314 kJ kmol\(^{-1}\) K\(^{-1}\)); \( T_a = \) absolute temperature (K).

**Thermodynamic properties (\( \Delta H, \Delta S, \Delta G \))**

The thermodynamic properties associated with the drying process were determined according to the method proposed by Jideani and Mpotokwana (2009). Arranged in Equations 10, 11, and 12 respectively, specific enthalpy, specific entropy, and Gibbs Free energy.

\[
\Delta H = E_a - R T_a \Delta H = E_a - R T_a \\
\Delta S = R \left( \ln D_0 - \ln \frac{K_s}{R_p} - \ln T_a \right) \Delta S = R \left( \ln D_0 - \ln \frac{K_s}{R_p} - \ln T_a \right) \\
\Delta G = \Delta H - T_a \Delta S \Delta G = \Delta H - T_a \Delta S
\]

where \( H = \) the specific enthalpy (J mol\(^{-1}\)); \( S = \) the specific entropy (J mol\(^{-1}\) K\(^{-1}\)); \( G = \) the Gibbs free energy (J mol\(^{-1}\)); \( K_b = \) the Boltzmann constant (1.38 x 10\(^{-23}\) J K\(^{-1}\)) and \( h_p = \) the Planck constant (6.626 x 10\(^{-34}\) J s\(^{-1}\)).

**Statistical analysis**

For the experimental adjustment of the drying kinetics, the computer program Statistic version 8.0 was used, using the non-linear regression analysis, by the Quasi-Newton method. The goodness of fit of the mathematical models to the observed statistical data was evaluated by the coefficient of determination (\( R^2 \)), chi-square (\( \chi^2 \)) and the root-mean-square error (RMSE, Root Mean Square Error).

**3. Results and Discussion**

Figure 1 shows the graph of the drying curves at temperatures of 40, 50, and 60 °C for penny royal leaves plotted with the values of the moisture ratio on a dry basis over time. It is possible to observe that with the increase of the drying air temperature, there is a reduction in the drying time. In a time of 360 min, for both temperatures, the quantification of UBS (kg of water/kg of dry mass) was 0.3093; 0.0190 and 0.0218 and the drying rate per min was 0.0026; 0.0009 and 0.0003 (kg of water/kg of dry mass).
Figure 1. Drying curves of the observed data of the drying kinetics of penny royal leaves, dried at temperatures of 40, 50, and 60 ºC until humidity lower than 10% in Ubu.

According to Fiorentin et al. (2010), this difference between drying times from 40 to 60 ºC occurs because at higher temperatures the sample starts to reduce its moisture more quickly at the beginning of drying, and therefore the drying time required will be shorter. The aforementioned effect is observed by several authors in their research as in the drying of plantains Milk et al. (2015), fermented grape pomace (Deamici et al., 2016), and strawberry drying (Oliveira, 2015).

With the drying process, the moisture contents found for each time were used to calculate the experimental moisture ratio (MR) values, which, in turn, were used to adjust the five chosen mathematical models (Table 1). The comparison between the observed moisture ratio (MR) values versus the MR predicted by the mathematical models is shown in Figures 2, 3, and 4. According to Corrêa et al. (2010), the figure was built to improve the range of adjustment of the models used to the data obtained in the drying, being able to affirm that the closer the experimental data are to the straight line, the greater the equality between the values. Analyzing the figures, it is possible to affirm that the experimental and predicted data of almost all models are well-adjusted.
Figure 2. Moisture ratio values were observed and predicted by the five mathematical models during drying of penny royal leaves at a temperature of 40 ºC.

![Graph showing moisture ratio values observed and predicted by five mathematical models at 40 ºC.](image)

Source: Authors.

Figure 3. Moisture ratio values were observed and predicted by the five mathematical models during drying of penny royal leaves at a temperature of 50 ºC.

![Graph showing moisture ratio values observed and predicted by five mathematical models at 50 ºC.](image)

Source: Authors.
Figure 4. Moisture ratio values observed and predicted by the five mathematical models during drying of penny royal leaves at a temperature of 60°C.

Table 2 presents the adjustment parameters of the approximate Diffusion, Two-Term, Logarithmic, Henderson & Pabis, Newton, and Page equations about the data collected during the drying kinetics of penny royal leaves. It can be seen that among the models tested at a temperature of 40 °C, the Approximate Diffusion model had the best fit to the collected data, for drying at 50 °C the Two-Term mathematical model and at a temperature of 60 °C the Logarithmic model, considering the highest values for R² and lowest values for x² and RMSE. It is noteworthy that models with R² ≥ 0.990 and RMSE < 0.1 can be considered as options to describe the mathematical modeling of drying kinetics.

Table 2. Coefficients of determination (R²), chi-square (x²), and root-mean-square error (RMSE) for the five mathematical models used to describe the drying process of penny royal leaves at 40, 50, and 60 °C.

| Nome do modelo       | 40 °C | 50 °C | 60 °C |
|----------------------|------|------|------|
|                      | R²   | x²   | RMSE | R²   | x²   | RMSE | R²   | x²   | RMSE |
| Difusão Aproximada   | 0.99658 | 0.00071 | 0.02860 | 0.99132 | 0.00260 | 0.05095 | 0.99207 | 0.00292 | 0.05408 |
| Two Terms            | 0.99192 | 0.00167 | 0.04087 | 0.99137 | 0.00258 | 0.05080 | 0.99182 | 0.00302 | 0.05491 |
| Logarítmico          | 0.98405 | 0.00165 | 0.04067 | 0.98532 | 0.00437 | 0.06614 | 0.99312 | 0.00254 | 0.05038 |
| Henderson & Pabis    | 0.99192 | 0.00167 | 0.04087 | 0.98525 | 0.00440 | 0.06630 | 0.99182 | 0.00302 | 0.05491 |
| Newton               | 0.98340 | 0.00342 | 0.05844 | 0.98207 | 0.00533 | 0.07304 | 0.99159 | 0.00310 | 0.05566 |
| Page                 | 0.99306 | 0.00143 | 0.03788 | 0.98953 | 0.00313 | 0.05593 | 0.99210 | 0.00291 | 0.05398 |

Source: Authors.

The coefficient of determination (R²) was high in all mathematical models tested in this work, above 98%, indicating quality in the adjustments. According to Madamba et al. (1996), these results indicate a satisfactory representation of the phenomenon under study, as the minimum value to have an acceptable reproduction of the models is R² greater than 95%. Similar scientific works, such as from Henriquez et al. (2014) that dry apple peels and Oliveira et al. (2020) who dehydrated cashew pulp at different temperatures and concentrations (*Brix), also found high coefficients of determination (> 99%) for their mathematical models.
The coefficients of the mathematical models are related to the drying temperature and the moisture content of the sample. According to Azevêdo et al. (2014), this phenomenon indicates that as drying temperatures increase, the speed at which water is removed from the sample is accelerated, an action attributed to the increase in the drying rate. That is, the values of \( k \) gradually increase as the temperature increases, as it is associated with the ease of removing moisture from the sample. This phenomenon can be easily seen in Table 3, where for all the models described, there was an increase in the values of gradual \( k \) for each drying temperature.

### Table 3. Drying constants (\( k \)) of the mathematical models analyzed during drying of pennyroyal leaves at 40, 50, and 60 °C.

| Name model          | 40 °C     | 50 °C     | 60 °C     |
|---------------------|-----------|-----------|-----------|
| Difusão Aproximada  | 0.14283   | 0.05746   | 0.50828   |
| Dois Termos         | \( k_1 \) = 0.00494 | \( k_1 \) = 0.007947 | \( k_1 \) = 0.01020 |
|                     | \( k_2 \) = 0.00446 | \( k_2 \) = 0.058716 | \( k_2 \) = 0.01024 |
| Logarítmico         | 0.004366  | 0.010226  | 0.00888   |
| Henderson & Pabis   | 0.00470   | 0.009841  | 0.01024   |
| Newton              | 0.00524   | 0.01090   | 0.00999   |
| Page                | 0.01438   | 0.02536   | 0.00754   |

Source: Authors.

The Logarithmic mathematical model presented the lowest drying constant (\( k \) min\(^{-1}\)) for the temperature of 40 °C in drying with airflow with an air velocity of 1.5 m/s (\( k = 0.004366 \)), followed by the Two Terms model for the temperature of 50 °C (\( k_1 = 0.00494 \)), and Page for the temperature of 60 °C (\( k = 0.00754 \)), thus, these models correspond to the lowest drying rates. For the Approximate Diffusion model, at the three drying temperatures, the highest values of \( k \) were found in comparison to the other models, and at 60 °C the maximum of \( k \) was reached (0.50828).

The effect indicates that with the increase in the temperature of the drying air, there was a decrease in the time needed for the penny royal leaves to reach the equilibrium water content.

Fick’s second law describes very well the dynamic behavior of the drying process during the period of decreasing moisture transfer rate over time, since effective diffusion (Def) is the main mass transfer mechanism (Henríquez et al., 2014). The increase in temperature directly affected the effective diffusion of the sample, as shown in Table 4, there was a decrease in Def when compared to temperatures of 40 and 60 °C.

### Table 4. Thermodynamic properties of pennyroyal leave specific enthalpy (\( \Delta H \)), specific entropy (\( \Delta S \)), Gibbs free energy (\( \Delta G \)), and effective diffusion (Def).

| Temperature (°C) | \( \Delta H \) (kJ mol\(^{-1}\)) | \( \Delta S \) (kJ mol\(^{-1}\) K\(^{-1}\)) | \( \Delta G \) (kJ mol\(^{-1}\)) | Def (m\(^2\) s\(^{-1}\)) |
|------------------|----------------------------------|-------------------------------------|----------------------------------|--------------------------|
| 40               | 45.0605                          | -0.1980                             | 107.0508                         | 2.925E+02                |
| 50               | 44.9774                          | -0.1982                             | 109.0316                         | 2.927E+02                |
| 60               | 44.8943                          | -0.1985                             | 111.0150                         | 2.929E+02                |

Source: Authors.

Regarding thermodynamic properties, Table 4, it can be seen that the specific enthalpy (\( \Delta H \)) decreased as the temperature used in the drying kinetics increased (40, 50 and 60 °C), confirming that the higher the temperature used, the less energy will be worn out during the drying process. On the other hand, specific entropy (\( \Delta S \)) and Gibbs free energy had a reverse
behavior to that of enthalpy, with an increase in values with increasing temperatures. The low entropy variation, between 0.3476 and 0.3481, is related to the low variation in the temperatures used (10°C), and negative values are usually related to changes in the material's structure.

4. Final Considerations

Among the drying models studied, the Logarithmic model and the Page model satisfactorily adjusted to the drying curves obtained experimentally for penny royal leaves. The drying temperature was influenced by the kinetics, and there was a decrease in Def when compared to temperatures of 40 and 60 °C. If we consider the most efficient time/temperature binomial for future use as conservation of penny royal leaves, the ideal for this type of plant material, sliced at 0.4 cm, would be at a temperature of 60 °C. For future work, the research group will use the data from the work to intensify the applicability of penny royal in food matrices.

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