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Post-harvest of parsley leaves (*Petroselinum crispum*): Mathematical modelling of drying and sorption processes

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Key words:
spice plant
processing
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ABSTRACT

Parsley is a species of wide production and trade in Brazil due to its high consumption as a condiment, fresh or dried. In the development of equipment used for drying, it is important to simulate and obtain theoretical information about the behavior of water loss for each product. Given the increasing use and potential commercialization of condimental plants, the objective of this work was to determine the isosteric heat of parsley leaves and fit mathematical models to the experimental data obtained in drying and desorption processes. The modified GAB and Midilli models were the most appropriate to describe the desorption isotherms and drying curves, respectively, for the studied temperatures. The isosteric heat varied from 3394.6 to 2830.0 kJ kg^{-1} for the equilibrium moisture content in the range from 0.0154 to 3.7232 (d.b.).

Palavras-chave:
planta condimentar
processamento
calor isostérico

Pós-colheita de folhas de salsinha (*Petroselinum crispum*): Modelagem matemática dos processos de secagem e dessorção

RESUMO

A salsinha é uma espécie de ampla produção e comércio no Brasil devido ao seu alto consumo como condimento in natura ou seco. No desenvolvimento de equipamentos para secagem é importante a simulação e a obtenção de informações teóricas a respeito do comportamento de cada produto durante a remoção de água. Tendo em vista a crescente utilização e o potencial de comercialização de plantas condimentares, neste trabalho objetivou-se determinar o calor isostérico das folhas de salsinha e ajustar modelos matemáticos aos dados experimentais da secagem e da dessorção. O modelo de GAB modificado e de Midilli foram os mais adequados para descrever as isotermas de dessorção e as curvas de secagem, respectivamente, para as temperaturas estudadas. O calor isostérico variou de 3394,6 a 2830,0 kJ kg^{-1} para o teor de água de equilíbrio na faixa de 0,0154 a 3,7232 (b.s.).

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Introduction

Parsley (*Petroselinum crispum* Mill.) is one of the vegetable species widely spread in Brazil and worldwide, and possibly the most universal of the condimental herbs (Lorenzi & Matos, 2008). Commonly in the Brazilian market, parsley leaves are commercialized as a condiment, both fresh and dried.

Vegetables are considered as highly perishable foods due to their high water content. Drying is a widely used method for preservation, increase of shelf life and reduction of size. Modeling assists in the analysis and understanding on the drying processes. Generally, model parameters are related to the required conditions, time and energy (Faal et al., 2015).

To minimize physical, chemical and microbiological alterations that occur after harvesting agricultural products, it is necessary to study the relationships between the product, relative air humidity and temperature through hygroscopicity, because they acquire the capacity to exchange gas, including water in form of vapor, with the surrounding environment. Studying equilibrium moisture content and water activity is important to design projects of post-harvest systems (Zeymer et al., 2017).

Once the hygroscopic equilibrium curves of a product are obtained, it is possible to determine its isosteric heat of sorption, a thermodynamic property of fundamental importance to compose mathematical models of drying and a parameter to estimate the minimum amount of heat required to remove certain amount of water (Costa et al., 2013). Given the above, this study aimed to perform mathematical modeling of desorption and drying of parsley leaves at different temperatures, and calculate the net isosteric heat of desorption.

Material and Methods

The study was carried out at the Laboratory of Post-harvest and Pre-processing of Agricultural Products, at the Engineering School of Volta Redonda, of the Fluminense Federal University (22° 30' 58.2" S; 44° 06' 16.5" W).

The plant material (parsley leaves) used in the experiment was provided by farmers from the southern region of Rio de Janeiro, Brazil. Initially, leaves were selected, cleaned from impurities and foreign materials, and homogenized.

In the desorption process, air temperatures of 20, 30, 40, 50 and 60 °C were tested in three replicates using an Aqualab Vapor Sorption Analyzer (VSA). To generate the isotherms, water activity is measured while a 0.005 g resolution scale weighs the sample. The device draws isotherms using the DDI method (Dynamic Dewpoint Isotherm), by constantly introducing dry air into the sample chamber, increasing or decreasing the relative moisture until it reaches the equilibrium. To predict the behavior of the desorption isotherm, these data were modeled using the mathematical models presented in Table 1.

The models were fitted to the experimental data through nonlinear regression analysis using the Quasi-Newton method. Models' degree of fit to the experimental data was evaluated based on the magnitude of the coefficient of determination (R^2), relative mean error (P), estimated mean error (SE) and root-mean-square error (RMSE), as described in Eqs. 11, 12 and 13 (Madamba et al., 1996; Afonso Júnior & Corrêa, 1999; Mohapatra & Rao, 2005).

Table 1. Mathematical models used to predict the desorption phenomenon in parsley leaves

Designation	Model	
Chung-Pfost	$X_e = a - b \,.\, \ln[-(T + c) \,.\, \ln(a_w)]$	(1)
Copace	$X_e = \exp[a - (b \,.\, T) + (c \,.\, a_w)]$	(2)
Modified GAB	$X_e = \dfrac{a \,.\, b \,.\, (c/T) .\, a_w}{\{[1 - b \,.\, a_w] \,.\, [1 - b \,.\, a_w + b \,.\, (c/T) .\, a_w]\}}$	(3)
Modified Halsey	$X_e = [\exp(a - b \,.\, T)/-\ln(a_w)]^{1/c}$	(4)
Henderson	$X_e = [\ln(1 - a_w)/-a \,.\, T_{abs}]^{1/c}$	(5)
Modified Henderson	$X_e = \{\ln(1 - a_w)/[-a \,.\, (T + b)]\}^{1/c}$	(6)
Sabbab	$X_e = a \,.\, \left(\dfrac{a_w^{\,b}}{T^c}\right)$	(7)
Sigma-Copace	$X_e = \exp[a - b.T + c.\exp(a_w)]$	(8)
Oswin	$X_e = {}^{(a - b.\, T)}\!/_{[(1 - a_w)/a_w]}{}^{1/c}$	(9)
Corrêa	$X_e = 1/(a.T^b + a_w^{\,c})$	(10)

X_e - Moisture content of the product, d.b.; a_w - Water activity, decimal; a, b, c - Parameters depending on the product; T - Air temperature, °C; T_{abs} - Absolute air temperature, K (Corrêa et al., 2002; Teixeira et al., 2012)

$$P = \frac{100}{n} \sum_{i=1}^{n} \frac{|Y - Y_0|}{Y} \qquad (11)$$

$$SE = \sqrt{\frac{\sum_{i=n}^{n} (Y - Y_0)^2}{DF}} \qquad (12)$$

$$RMSE = \sqrt{\frac{1}{n} \sum_{i=1}^{n} (Y - Y_0)^2} \qquad (13)$$

where:

n - number of observations;
Y - experimental value;
Y_0 - value estimated by the model; and,
DF - degrees of freedom of the model.

Net isosteric heat of desorption (q_{st}) was obtained by the Clausius-Clapeyron equation (Eq. 14), modified by Wang & Brennan (1991). Water activity, temperature and equilibrium moisture content were obtained from desorption isotherms of the product, using the model of best fit to the observed data. Integral isosteric heat of desorption was calculated using Eq. 15. The latent heat of vaporization of free water (L) was determined by Eq. 16.

$$\mathrm{In}(a_w) = -\frac{q_{st}}{R} \cdot \frac{1}{T_{abs}} + C \qquad (14)$$

$$q_{st} = A.\exp(-B.X_e) \qquad (15)$$

$$L = 2502.2 - 2.39 \cdot T_m \qquad (16)$$

where:

q_{st} - net isosteric heat of desorption, kJ kg^{-1};

R - universal gas constant, for water vapor, 0.4619 kg^{-1} K^{-1};
a_w - water activity, decimal;
T_{abs} - absolute temperature, K;
C - model coefficient;
A and B - fit parameters;
X_e - equilibrium moisture content expressed on dry basis, decimal;
L - latent heat of vaporization of free water, kJ kg^{-1}; and,
T_m - mean temperature in the studied range, in °C.

To evaluate the drying process, parsley leaves were separated into two groups: Group 1, in which leaves were not subjected to any sanitation process, and Group 2, in which samples were washed in running water, immersed for 15 min in a sanitizing solution of 200-ppm sodium hypochlorite and rinsed twice with distilled water (Costa et al., 2012). Before weighing, to perform the drying process, the superficial excess of water was removed in a manual centrifuge for vegetables.

Different temperatures of the drying air (40, 50 and 60 °C) were used in a tray dryer equipped with an electric air-heating system. To obtain the drying curves, periodic weighings were performed until reaching the final moisture content of 0.149 (d.b.). This value was fixed as an average between the values established for medicinal plants in different Pharmacopoeias of various countries, which range between 0.087 and 0.163 (d.b.) (Simões et al., 2010). The experiment was carried out in three replicates. The moisture content of the samples was determined by the method ASAE (2000), in three replicates. Drying curves were fitted using Eqs. 17 to 29, presented in Table 2.

Table 2. Mathematical models used to predict the drying phenomenon

Designation	Model	
Approximation of diffusion	$RX = a.\exp(-k.t) + (1-a).\exp(-k.b.t)$	(17)
Two terms	$RX = a.\exp(-k_0.t) + b.\exp(-k_1.t)$	(18)
Two-term exponential	$RX = a.\exp(-k.t) + (1-a).\exp(-k.a.t)$	(19)
Henderson-Pabis	$RX = a.\exp(-k.t)$	(20)
Modified Henderson-Pabis	$RX = a.\ \exp(-k.t) + b.\exp(-k_0.t) + c.\exp(-k_1.t)$	(21)
Logarithmic	$RX = a.\exp(-k.t) + b$	(22)
Midilli	$RX = a.\exp(-k.t^n) + b.t$	(23)
Newton	$RX = \exp\ (-k.t)$	(24)
Page	$RX = \exp\ (-k.t^n)$	(25)
Modified Page	$RX = \exp[-(k.t)^n]$	(26)
Thompson	$RX = \left[\frac{-a-(a^2+4.b.t)^{1/2}}{2.b}\right]$	(27)
Verna	$RX = a.\exp(-k.t) + (1-a).\exp(-k_1.t)$	(28)
Wang-Sing	$RX = 1 + a.t + b.t^2$	(29)

RX - Moisture content ratio; t - Time; a, b, c, k, k_o, k_1, n - Model's regression fit terms (Madamba et al., 1996; Doymaz, 2004; Mohapatra & Rao, 2005)

Moisture content ratio (RX) was calculated using Eq. 30. The hygroscopic equilibrium moisture content obtained in the modeling of the desorption curves was used.

$$RX = \frac{X - X}{X - X_e} \tag{30}$$

where:

RX - moisture content ratio of the product (dimensionless);
X - moisture content of the product, decimal (d.b.);
X_e - equilibrium moisture content of the product, decimal (d.b.); and,
X_i - initial moisture content of the product, decimal (d.b.).

Models' degree of fit to the experimental data was evaluated as described for the desorption isotherms.

RESULTS AND DISCUSSION

Among the evaluated models, Modified GAB showed the best fit to the observed data of equilibrium moisture content as a function of water activity of parsley leaves, with the highest coefficient of determination (96.35%) and lowest values of root-mean-square error (0.3369) and estimated mean error (0.3383), justifying the choice of this model to represent parsley desorption curves. Similar results were found by Edoun et al. (2010), who determined the equilibrium moisture content of eru (*Gnetum africanum*) leaves for the temperature range from 30 to 50 °C, and observed that the GAB model was the one that best described the desorption isotherms.

The estimated parameters of the Modified GAB model were: b = 0.9133 and c = 2.6697. The parameter "b" of the Modified GAB model is called correction factor, because it corrects the properties of the multilayer water molecules. When its value is close to 1, there is virtually no difference between multilayer molecules and liquid molecules, i.e., the vaporization heat is close to the water vaporization heat. The parameter "c" is related to the differences of chemical potential between the monolayer and upper layers, representing the binding energy between water molecules in the monolayer and sorption sites on the product's surface. The higher its value, the more strongly the water is bound to the monolayer (Basptestini et al., 2017).

According to the classification of IUPAC (1985), the desorption isotherms of parsley leaves exhibit a type-II shape, sigmoid, due to the results obtained for the parameters "b" and "c" of the Modified GAB model, which indicate the type of isotherm, showing values from 0 to 1.0 for "b" and higher than 2.0 for "c". This is a typical shape observed in plants, such as wormseed (*Chenopodium ambrosioides*) leaves by Jamali et al. (2006).

The experimental data of equilibrium moisture content as a function of water activity of parsley leaves (desorption isotherms), at the studied temperatures, and the curves fitted by the Modified GAB model are presented in Figure 1. Equilibrium moisture content decreases with the reduction in water activity for the studied temperatures, which is consistent with most hygroscopic products. Such behavior was observed by Silva et al. (2007) for pot marigold (*Calendula officinalis* L.).

The Modified GAB model was used to analyze the isosteric heat of desorption because it showed the best fit to the desorption isotherms. The integral isosteric heat of desorption of the leaves with moisture contents between 0.0154 and 3.7232 (d.b.) ranged from 3394.6 to 2483.0 kJ kg^{-1} (Figure 2). As the equilibrium moisture content decreases, there is an increase in the integral isosteric heat (Q_{st}); the lower the moisture content in parsley leaves, the more energy is required to evaporate

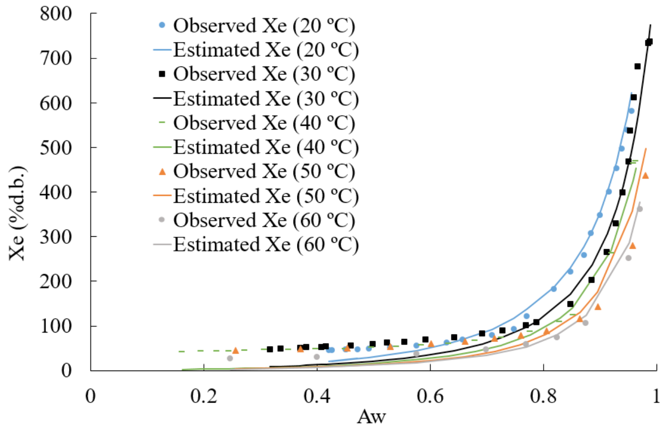

Figure 1. Desorption curve of parsley leaves fitted by the Modified GAB model at different temperatures

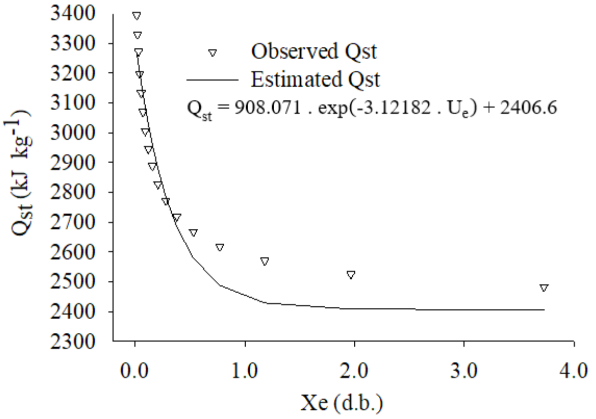

Figure 2. Experimental and estimated values of integral isosteric heat of desorption as a function of the equilibrium moisture content of parsley leaves

water bound to the product's biological structure. These results agree with those of Mohamed et al. (2005) and Ghodake et al. (2007), who studied sorption isotherms of dry leaves of black tea (*Camellia sinensis* L.) and in leaves of orange (*Citrus aurantium*), respectively.

The parameters estimated for the Exponential model were: A = 908.071 and B = 3.12182, with a coefficient of determination of 91.63%, which mathematically led to the equation of integral isosteric heat of desorption of parsley leaves as a function of equilibrium moisture content and mean temperatures evaluated. An inverse proportionality was observed between desorption heat and equilibrium moisture content (Figure 2), which is consistent with Iglesias & Chirife (1976), who attributed such behavior to the water sorption process. Other agricultural products showed the same behavior, such as pulp and peel of pineapple (*Ananas comosus*) (Teixeira et al., 2012) and leaves of spearmint (*Mentha* sp.) (Ayadi et al., 2014).

Among the models evaluated in the prediction of parsley drying from Groups 1 and 2, Midilli showed the best fit to the observed moisture content data for both groups. While the values of SE, RMSE and P (%) varied from 0.0027 to 0.594, 0.022 to 0.561 and 1.25 to 35.3, respectively, for all models evaluated, the Midilli model in both groups showed lower SE and RMSE, ranging from 0.0026 to 0.0075 and 0.0023 to 0.0054, respectively.

The value of P varied between 1.25 and 7.11%, lower than 10%, considered as good fit according to Mohapatra & Rao (2005). The model also showed high R^2, above 99.95% at all temperatures, which according to Madamba et al. (1996) indicates a good fit to represent the drying phenomenon.

In studies with other plant species, the Midilli model was the one that best fitted to the experimental data of drying, as observed by Ayadi et al. (2014) in the convective drying of spearmint (*Mentha* sp.) at temperatures of 40, 45, 50 and 55 °C, and by Radünz et al. (2010) in the drying of leaves of guaco (*Salvia officinalis*) using tray dryers at temperatures of 40, 50, 60, 70, 80 and 90 °C. This model was developed by Midilli et al. (2002) to describe the single-layer drying kinetics and statistically compared with other empirical or semi-empirical models commonly used. The drying kinetics experiments were conducted using mushrooms, pollen and pistachio, which were dried under laboratory conditions, direct sunlight, and in forced-air sunlight system.

Table 3 shows the Midilli model parameters for parsley drying in the different groups. The results corroborate those found by Goneli et al. (2014) in the drying of aroeira leaves in tray dryer at temperatures from 40 to 70 °C.

The Midilli model parameters "a" and "b" are related to the mathematical fits associated with factors external to the drying, since the model is semi-empirical. According to Madamba et al. (1996), the drying constant "k" can be used as an approximation to characterize the effect of drying air temperature and is related to the effective diffusivity in the drying process in the decreasing phase, and the liquid diffusion controls the drying process. The drying constant "k" (Table 3) increases with the increment in drying air temperature for both groups, representing the effect of external drying conditions, such as air temperature and relative humidity.

For the "n" value, the results evidenced increase with the increment in drying air temperature. This parameter depends upon the product and drying conditions, reflecting the extension of the product's internal resistance to drying, for certain conditions, thus explaining the moisture gradients established inside the product during the process at the rate it occurs (Misra & Brooker, 1980).

According to Figure 3, drying time decreases with the increment in air temperature. The times required to complete the drying process were 48, 28.5 and 15.5 h, in Group 1, and 31, 19 and 12 h, in Group 2 for the temperatures of 40, 50 and 60 °C, respectively. In addition, drying temperature exerts influence on parsley drying speed. Similar results have been found in the drying of leaves of thyme (Rocha et al., 2012) and

Table 3. Midilli model parameters for parsley drying obtained in the different temperatures

Groups	Temp. (°C)	Model parameters			
		a	b	K	N
1	40	1.0034	-0.0007	0.0908	0.8741
	50	0.9993	-0.0014	0.1350	0.8782
	60	0.9916	-0.0024	0.2377	0.8925
2	40	0.9981	-0.0008	0.1451	0.9058
	50	0.9986	-0.0014	0.1910	0.9495
	60	1.0009	-0.0033	0.2641	0.9501

1 - Without sanitation; 2 - With sanitation

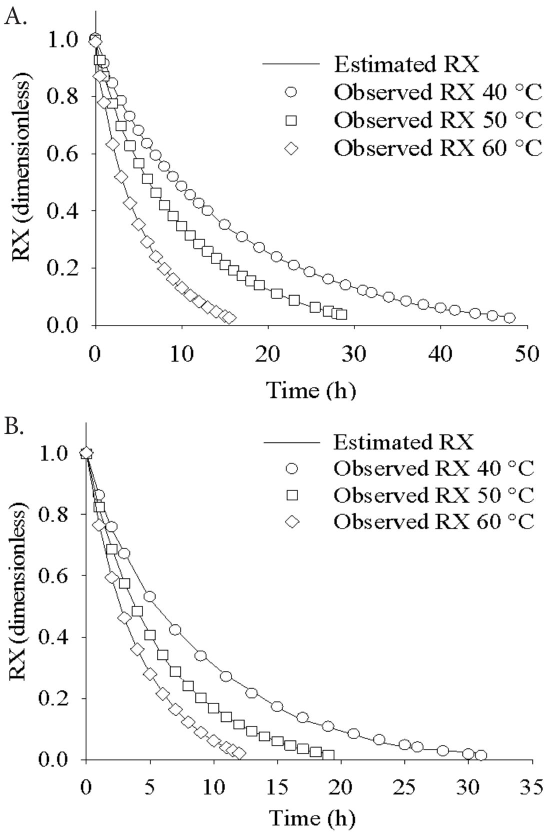

Figure 3. Moisture content ratio, experimental and estimated by the Midilli model, in the drying of parsley leaves at the evaluated temperatures: Group 1 (A) and Group 2 (B)

guaco (*Salvia officinalis*) (Radünz et al., 2010). Comparing the drying time between the studied processes, it was observed that, after sanitation, when leaves were manually centrifuged, there was a reduction of about 30% in the time. This fact may be related to the cell restructuration after sanitation.

Conclusions

1. Modified GAB and Midilli models were the most appropriate to describe the desorption curves and drying kinetics of parsley leaves, respectively, in the studied temperature range.

2. The desorption isotherms exhibit a type-II shape, sigmoid, and the equilibrium moisture content decreased with the reduction in water activity for the studied temperatures.

3. Manual centrifugation of parsley leaves reduces the drying time by approximately 30%.

4. Isosteric heat varied from 3394.6 to 2830.0 kJ kg^{-1} for equilibrium moisture content in the range from 0.0154 to 3.7232 (d.b.).

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