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Research Article

Citron melon peel flours: drying kinetics and physicochemical evaluation

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Abstract

In the utilization of citron melon (*Citrullus lanatus* var. *citroides*), it is necessary to deal with a large amount of residues constituted by the peels. These materials commonly discarded can be fully utilized, since they are a source of nutrients. The peels contain not only fibers, but also proteins, sugars and minerals, which, after drying, are concentrated to values that make them interesting for various uses, including for the enrichment of flours or combinations to prepare bakery products. Therefore, the drying of the peels, besides enabling the conservation and storage at room temperature, expands the forms of use and the possibilities of entering the production chain. This study aimed to characterize physico-chemically and determine drying kinetics, effective diffusivity, activation energy and thermodynamic properties of citron melon peels at different drying temperatures. The peels were dried in an oven with forced air circulation, in a thin layer, at temperatures of 60, 70, 80 and 90 °C. With the data collected in the drying, the drying kinetic curves were constructed and eleven mathematical models were fitted to the experimental data. The dried material was crushed in a knife mill and characterized for physicochemical parameters. Midilli model resulted in the best fits, followed by Page and Approximation of Diffusion models. Effective diffusivity increased with drying temperature; activation energy was obtained from the Arrhenius equation and was equal to 8.18 kJ/mol. Enthalpy and entropy were reduced with increased temperature, while Gibbs free energy increased.

Keywords: agricultural waste; *Citrullus lanatus* var. *citroides*; environment; sustainability; thermodynamic properties

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Introduction

Citron melon (*Citrullus lanatus* var. *citroides*), commonly known as fodder melon, red-seeded melon or Kalahari melon, has percentages of fat and crude fiber comparable to those of forage plants such as prickly pear (Mujaju *et al*., 2010; Jesen *et al*., 2011; Mustafa and Alamin, 2012). Native to Africa, it can achieve considerable yields, being used either fresh or as bran in animal feed. However, little is known about the chemical composition of this fruit, in addition to pulp, peel and seeds, information that is essential to understand the potentials of the crop, including for possible use in human diet. Depending on the fruit, the main residues generated during processing are: peel, seeds and bagasse (Okino-Delgado *et al*., 2018; Ueda *et al*., 2022). These residues, when utilized, serve mostly for animal feed; however, they could be used to obtain new food products, mainly due to their low cost and for containing vitamins, minerals, fibers and antioxidant compounds that are important for physiological functions, and their use may contribute to improving the nutritional value of the diet of populations and reducing agro-industrial residues (Zabalaga *et al*., 2016; Souza and Vieira, 2020).

The high moisture content found in agro-industrial residues makes them highly perishable, so it is necessary to search for methods that extend their useful life. The simplest method to achieve this purpose is drying, whose advantage is to enable the transformation of the dry product into flour, a useful form of presentation to make up mixtures for nutritional enrichment, improvement in acceptability and reduction of costs with raw materials. Drying is reduction the moisture to safe levels for the storage operation, preserving physical and chemical characteristics, ensuring the use of the product in periods when the fruit is not available (Resende *et al*., 2018; Venturin and Silva, 2019; Barros *et al*., 2020). For the design and optimization of drying equipment, it is important to determine the drying kinetics, fit mathematical models to the data and select those that result in the best prediction of water loss, based on the experimental data obtained (Costa *et al*., 2015), justifying the importance of theoretical information on the drying of each raw material, especially those of biological origin, which are characterized by high physical and chemical variability.

The utilization of agro-industrial residues is an alternative that makes it possible to increase the income of the producer and reduce losses associated with production. Their chemical characteristics and applications have been studied by several authors, such as Cazarin *et al*. (2014), studying the antioxidant capacity and chemical composition of *Passiflora edulis*; Queiroz *et al*. (2015), evaluating the chemical and phytochemical composition of *Litchi chinensi*) peel and seed flours; Bender *et al*. (2016), in a study on the characterization of *vinifera* L. skin flour and its use in extruded snack; Santos *et al*. (2017), who performed physical and chemical analyzing of the dry peels of *Cereus undatus* Haworth; Rybka *et al*. (2018), assessing the composition of peel flour from different *Mangifera indica* cultivars; Almeida *et al*. (2020b), who determined bioactive compounds and performed physicochemical composition of *Plinia cauliflora* peel flour obtained by convective drying and freeze-drying.

In view of the above, this study aimed to physicochemically characterize and determine the behaviour of drying kinetics, in addition to effective diffusivity, activation energy and thermodynamic properties of citron melon peel subjected to different drying temperatures.

Materials and Methods

Raw material processing and physic-chemical analysis

The raw material used was the peel of citron melon (*Citrullus lanatus* var. *citroides*). Citron melon fruits, acquired in the municipality of Areial, State of Paraíba, Brazil, around the geographic coordinates of 7° 02’ 52” S latitude, 35° 55’ 51” W longitude and altitude of 695 m, were harvested at the mature stage and transported to the laboratory for processing.

The fruits were washed under running water and then immersed in sodium hypochlorite solution at 100 ppm for 30 min. The citron melons were fractionated into smaller parts and the pulp was manually separated from the peel. The peels were washed under running water and placed in trays for evaporation of surface water. Subsequently, the peels were cut with a stainless-steel knife in rectangular shape with average thickness of 3.25 mm.

For the fresh peels of citron melon and for the flours, the following analyses were performed in triplicate, according to the analytical procedures of the Adolfo Lutz Institute (IAL, 2008): moisture content, by the gravimetric method in an oven at 105 °C, until reaching constant mass; ash content, by incineration in muffle furnace at 550 °C, with results expressed in percentage (w/w); total acidity, by titrimetry with 0.1 mol/L NaOH; pH, determined with pH meter previously calibrated with pH buffer solutions 7.0 and 4.0; and water activity (a_w), determined in an Aqualab meter (3TE, Decagon Devices) at 25 °C. The amount of lipids was determined by extraction with cold solvent mixture, according to the method described by Bligh and Dyer (1959). According to AOAC (2016), the protein content was calculated by the total amount of nitrogen in the product multiplied by the protein conversion factor of 6.25 and expressed in g/100 g. Starch content was determined by the anthrone method (Stevens and Chapman, 1955). Total sugars were determined by the methodology described by Yemm and Willis (1954), in which the samples were analysed in spectrophotometer at 620 nm and the quantification of sugars was based on the standard glucose curve. Reducing sugars were determined by methodology of Miller (1959), using 3,5-dinitrosalicylic acid (DNS) as an oxidizing agent, with readings in spectrophotometer (Coleman, 3.5 D, Santo André, SP, Brazil) at 540 nm. Non-reducing sugars were determined by the difference between the values of total sugars and reducing sugars.

Drying kinetics of the peels

Citron melon peels were distributed in screened baskets (Figure 1a) and dried in a forced air circulation oven (Figure 1b), varying the drying temperature (60, 70, 80 and 90 °C) and with air speed of 1.0 m/s. Drying kinetics was determined by weighing the baskets with the samples at regular intervals of 5, 10, 20, 30 and 60 min, until the equilibrium was reached, and determining the dry mass in an oven at 105 °C, according to the methodology of the Adolfo Lutz Institute (IAL, 2008). The dried material (Figure 1c) was crushed in a knife mill and the flours (Figure 1d) obtained were characterized according to the physicochemical parameters. With the experimental data of drying kinetics, the moisture content ratios of the samples were calculated according to Equation 1.

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

Where: RX - moisture content ratio of the sample (dimensionless); X - moisture content of the sample at a given drying time (d.b.); X_i - initial moisture content of the sample (d.b.); X_e - equilibrium moisture content of the sample (d.b.).

The drying rates of citron melon peel at temperatures of 60, 70, 80 and 90 °C were determined from Equation 2, considering the moisture contents (d.b.) in each interval.

$$TX = \frac{X_{t+dt} - X_t}{dt} \tag{2}$$

Where: TX - drying rate (kg/kg/min); X_{t+dt} - moisture content in t + dt (kg water/kg dry matter); X_t - moisture content at a specific time (d.b.); dt - time interval between two consecutive measurements; and t - time (min).

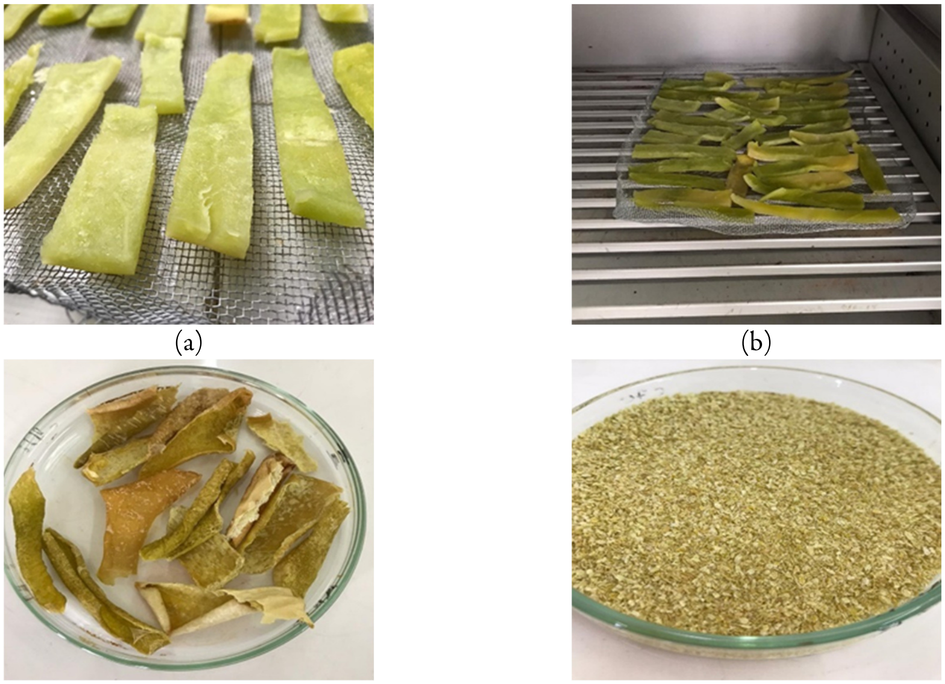
(a) (b) (c) (d)

Figure 1. Citron melon peel; (a) Peel slices; (b) Drying of slices; (c) Dehydrated peels; and (d) Citron melon peel flour

With the data collected, the mathematical models (Table 1) of Newton, Modified Page, Page, Henderson and Pabis, Two-Term Exponential, Wang and Singh, Logarithmic, Exponential Logarithmic, Approximation of Diffusion, Two Terms and Midilli were fitted to the experimental data, through nonlinear regression analysis, by the Quasi-Newton method, using the computer program Statistica 7.7®.

Table 1. Mathematical models fitted to drying kinetics data

Model designation	Models	Equation
Newton	$RX= \exp(-k.t)$	(3)
Modified Page	$RX= \exp[-(k.t)^n]$	(4)
Page	$RX= \exp(-k.t^n)$	(5)
Henderson and Pabis	$RX= a \exp(-k.t)$	(6)
Two-Term Exponential	$RX= a.\exp(-k.t) +(1-a)\exp(-k.a.t)$	(7)
Wang and Singh	$RX = 1 + a.t + b.t^2$	(8)
Logarithmic	$RX= a \exp(-k.t) +c$	(9)
Exponential Logarithmic	$RX= a.\exp(k0.t) + (1-a) \exp(-k.a.t)$	(10)
Approximation of Diffusion	$RX= a.\exp(-k.t)+(1-a).\exp(-k.b.t)$	(11)
Two Terms	$RX= a. \exp(-k_0.t) +b.\exp(-k_1.t)$	(12)
Midilli	$RX= a. \exp(-k.t^n) +b.t$	(13)

Where: RX - moisture content ratio; k - drying constant; a, b, c, n, q - coefficients of the models; t - drying time (min).

Coefficient of determination (R^2), mean squared deviation (MSD) and chi-square ($\chi 2$), according to Equations 14, 15 and 16, respectively, were used as criteria for the fit of the mathematical models to the experimental data of citron melon peel drying.

$$R^2 = 1 - \left(\frac{\sum_{i=1}^{n}(RX_{pred,i} - RX_{exp,i})^2}{\sum_{i=1}^{n}\left(RX_{exp,i} - RX_{pred,i}\right)^2}\right) \tag{14}$$

$$MSD = \left[\frac{1}{n}\sum_{i=1}^{n}(RX_{pred,i} - RX_{exp,i})^2\right]^{\frac{1}{2}} \tag{15}$$

$$\chi^2 = \frac{1}{n-N}\sum_{i=1}^{n}(RX_{exp,i} - RX_{pred,i})^2 \tag{16}$$

Where: R^2 - coefficient of determination; MSD - mean squared deviation; χ^2 - chi-square; $RX_{pred,i}$ - moisture content ratio predicted by the model; $RX_{exp,i}$ - experimental water ratio content; n - number of observations; N - number of model constants.

The effective diffusivity of watermelon peels, at temperatures of 60, 70, 80 and 90 °C, was calculated according to the analytical solution of Fick's second law (Equation 17), which is based on the theory of liquid diffusion, considering the geometric shape of the peels similar to that of an infinite slab (average peel thickness of 0.00325 m), with three-term approximation. It was considered that the distribution of the initial moisture content was uniform, the resistance to external mass transfer, the temperature gradient within the sample and the shrinkage were negligible, and the effective diffusivity was constant for a given drying temperature.

$$RX = \frac{X - X_e}{X_I - X_e} = \frac{8}{\pi^2}\sum_{n=0}^{\infty}\frac{1}{(2n+1)^2}\exp\left[-(2n+1)^2\pi^2 D_{ef}\frac{t}{L^2}\right] \tag{17}$$

Where: D_{ef} - effective diffusivity (m^2/s); n - number of terms of the equation; L - layer thickness (m); t - time (s).

The effect of drying temperature on the effective diffusivity of citron melon peel was evaluated using the Arrhenius equation (Equation 18).

$$D_{ef} = D_{ef0}\exp\left(-\frac{E_a}{RT}\right) \tag{18}$$

Where: D_{ef} - effective diffusivity (m^2/s); D_{ef0} - pre-exponential factor (1/s); Ea - activation energy (kJ/mol); R - universal constant of gases (0.008314 kJ/mol K); T - absolute temperature (K).

The thermodynamic properties (enthalpy, entropy and Gibbs free energy) of the citron melon peel drying process at temperatures of 60, 70, 80 and 90 °C were quantified using the method described by Jideani and Mpotokwana (2009), respectively according to Equations 19, 20 and 21.

$$\Delta H = E_a - RT \tag{19}$$

$$\Delta S = R\left[\ln(Deff_o) - \ln\left(\frac{k_B}{h_p}\right) - \ln(T)\right] \tag{20}$$

$$\Delta G = \Delta H - T\Delta S \tag{21}$$

Where: ΔH - specific enthalpy (kJ/mol); ΔS - specific entropy (kJ/mol K); ΔG - Gibbs free energy (kJ/mol); K_B - Boltzmann constant (1.38 × 10-23 J/K); hp - Planck constant (6.626 × 10-23 J/s); T - absolute temperature (K).

The results obtained in the characterization of fresh peels and flours were evaluated using a completely randomized design, composed of 5 treatments (fresh samples and samples dried at 60, 70, 80 and 90 °C) and 3 replicates. The data were subjected to analysis of variance (ANOVA) and the means compared by Tukey test at 5% probability level using Assistat 7.7 software (Silva and Azevedo, 2016).

Results and Discussion

Table 2 shows the mean values obtained in the physicochemical characterization of fresh citron melon peel and flours obtained after drying the peels at temperatures of 60, 70, 80 and 90 °C. The values of fresh samples and flours differed statistically for all the parameters evaluated. The values referring to moisture content, at a level conducive to 60 °C, gradually decreasing with the increase up to 90 °C. The moisture content is in accordance with the value recommended by the Brazilian Legislation (Brasil, 2005) for products dried and processed in the form of flour, which is at most 15%. Similar results were found by Vieira *et al.* (2017) for melon peel flour (9.86%) and by Bender *et al.* (2016) in the characterization of grape skin flour (7.17%). Gonçalves *et* *al*. (2016), when drying green banana peels at different drying temperatures, obtained samples with higher moisture contents of 14.60 and 14.54% at drying temperatures of 65 and 75 °C, respectively.

Water activity (aw), above 0.990 in the fresh sample, decreased with dehydration, and the gradual increase in drying temperature led to consequent and statistically significant reductions among all flours. In addition, all flours had a_w value suitable for storage under safe conditions ($a_w < 0.6$). Similarly, aw value below 0.6 ($a_w = 0.430$) was found by Cazarin *et al*. (2014) for passion fruit peel flour dehydrated at temperature of 50 °C.

Table 2. Physicochemical characterization of fresh citron melon peel and flours obtained at drying temperatures of 60, 70, 80 and 90 °C

Parameters	In natura	60 °C	70 °C	80 °C	90 °C
Moisture content (%)	93.69±0.88a	8.61±0.26b	5.70±0.17c	4.40±0.19d	3.33±0.10d
Water activity (a_w)	0.994±0.001a	0.458±0.002b	0.323±0.002c	0.254±0.004d	0.224±0.004e
Ash (%)	8.56±0.09a	6.90±0.38bc	6.71±0.57c	7.59±0.12b	7.58±0.04b
Total titratable acidity (%)	0.91±0.01c	0.99±0.01a	0.97±0.01b	0.99±0.00a	0.95±0.01b
pH	5.38±0.03a	5.00±0.04b	4.93±0.01c	4.86±0.02d	4.83±0.01d
Lipid (%)	9.61±0.24a	2.56±0.06b	2.35±0.09b	2.42±0.05b	2.24±0.11b
Protein (%)	1.54±0.03c	2.23±0.25ab	2.69±0.24a	2.14±0.04b	2.03±0.24bc
Starch (%)	1.76±0.11d	4.43±0.16a	3.40±0.05b	2.38±0.06c	1.85±0.09d
Total sugars (g/100 g)	8.82±0.08e	26.22±0.42d	27.86±0.17c	41.69±0.09a	38.50±0.00b
Content of reducing sugars (g/100 g)	3.48±0.017a	0.68±0.00c	0.66±0.00c	0.63±0.00d	0.70±0.00b
Non-reducing sugars (g/100 g)	5.07±0.06e	25.53±0.47d	27.20±0.05c	41.05±0.10a	37.79±0.00b

The ash contents, similar to those found in the fresh peel of other vegetables, showed a reduction resulting from drying. Ash contents in the dried samples were different from those reported in several other studies with similar materials. A study evaluating the ash content of umbu and umbu-cajá peel flours reported values of 3.10 and 3.88%, respectively (Silva *et al*., 2014). Mallek-Ayadi *et al*. (2017) and Gómez-García *et al*. (2021) studied melon (*Cucumis melo* L.) peels and found ash contents of 4.41 and 8.44%, respectively.

There was an increase in total titratable acidity with the drying process, and it was not possible to identify the effect of drying temperature differences. The legislation recommends an acidity limit of 2% for flours, so the results obtained here (0.95-0.99% d.b.) meet the current legislation (Brasil, 2005). Nunes *et al*. (2017), studying pineapple residues (peels and stems), found total titratable acidity values of 1.74, 1.89, and 2.16% for temperatures of 50, 60 and 70 °C, respectively.

Inversely to acidity, drying reduced the pH of the flours compared to the fresh sample, and the temperature increase gradually reduced the pH between subsequent temperatures, probably due to the degradation of organic acids. Santos *et al*. (2017), dehydrated pitaya (*Hylocereus guatemalensis)* peel and found acidity values of 2.82, 2.68 and 2.35% citric acid for temperatures of 50, 60 and 70 °C, respectively, which corresponded to pH values of 5.06, 5.09 and 5.13, the samples were above the range considered safe to maintain microbiological stability (pH < 4.5), excluding other factors such as moisture content and water activity.

The lipid content, which was not higher in the fresh peel, decreased in the dehydrated samples, because the thermal processing reduced it, suppressing the lipases. The increase in drying temperature did not lead increases in lipid content, which was between 2.24 and 2.56% d.b., for temperatures of 90 and 60 °C, thus diverging from any effect of concentration. Leão *et al*. (2017), when evaluating the chemical composition of pequi (*Caryocar brasiliense*) peel flour, found lipid contents of 0.34% (epicarp + mesocarp) and 0.18% (mesocarp).

Regarding protein content, the value increased with the drying process. Leão *et al.* (2017) found a value of 3.25%, for the pequi peel flour. Amarante *et al.* (2018) and Almeida *et al.* (2020a) studied the chemical properties of feijoa (*Acca sellowiana*) peel flour and found values of 0.80 and 1.42%, respectively. Zhu (2018) cited that differences in protein content can be explained by several factors, such as genetic variation, differences of climate, temperature and conditions, as well as drying and processing conditions.

With the dehydration process, there was a decrease in starch content for citron melon peel flours, compared to the fresh peel. Probably, this behaviour is caused by the degradation of starch with the drying process. Brazilian food legislation (Anvisa, 2003) mentions that, for starch to be marketed and used in food, it must have at least 84% starch. Thus, the starch content obtained in the peel flours was low, compared to the ideal, with higher levels of other constituents, not being ideal for products that require gelatinization.

Total sugars in the flours increased compared to the fresh sample and in general with the increase in dehydration temperature, except for flour dried at 90 °C. This increase is probably related to the heat treatment employed and the increase in TTA. The content of reducing sugars determined in the fresh sample decreased in the flours, as an effect of dehydration. This behaviour confirms the consumption of reducing sugars during the drying process, under the temperature conditions used, which can be attributed to the Maillard reaction. Although the highest value was determined at 90 °C, there is no trend of direct variations with temperature. Almeida *et al.* (2020b) applied conservation techniques for jaboticaba peels and found values of reducing sugars of 25.24 and 26.29% for convective drying and freeze-drying, respectively. The authors add that the higher the sugar content, the greater the hygroscopicity of the product. This is of practical importance, especially with regard to the conditions of marketing of dehydrated products. In relation to non-reducing sugars, there was an increase with the increase in temperature.

The drying rates of citron melon peels at temperatures 60, 70, 80 and 90 °C are shown in Figure 2. An approximate behaviour of constant drying rate is observed in the first minutes of the process, a period in which the average drying rate is higher, a behaviour explained by the loss of surface water from the peel, which in longer times decreases as the internal mass transfer processes become preponderant. Silva *et al.* (2017a) reported that the increase in drying air temperature promotes greater action of the diffusive mechanisms inside the sample, resulting in a greater amount of active sites on the evaporation surface, hence leading to a higher rate of water removal from the product and reducing drying time.

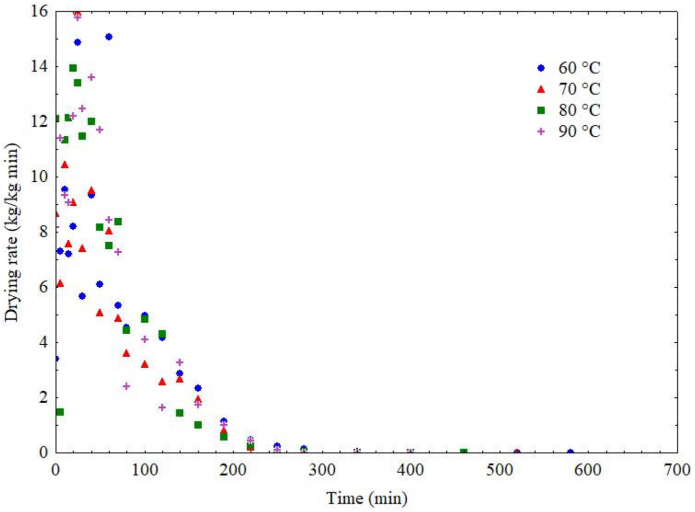

Figure 2. Drying rates of citron melon peels at temperatures of 60, 70, 80 and 90 °C

Martins *et al.* (2014), when drying mulungu bark at temperatures of 40, 50, 60 and 70 °C, found that the drying processes were performed in falling rate periods, with higher rates at the beginning and reduction over time. The authors found the highest drying rate at 70 °C, with magnitude greater than 9×10^{-3} kg/min, while the lowest drying rate was verified at 40 °C (value greater than 2×10^{-3} kg/min).

Table 3 presents the parameters of the models fitted to the drying kinetics data of the citron melon peels at temperatures of 60, 70, 80 and 90 °C, with statistical results of the fits of the models analysed for reliability, adequacy and consistency according to Omolola *et al.* (2019), by means of the coefficients of determination (R^2), mean squared deviations (MSD) and chi-squares (χ^2).

It can be observed that all models, except Wang and Singh, showed R^2 values higher than 0.970, indicating that they are adequate for predicting the drying kinetics of the material. Martins *et al.* (2015), Alexandre *et al.* (2019) and Santos *et al.* (2020), stated that $R^2 > 0.95$ indicates a good representation of the results through mathematical models, but evaluating only this parameter does not constitute a good criterion for selecting nonlinear models, hence requiring joint evaluation of other statistical parameters such as MSD and χ^2.

MSD ranged from 0.0092 (Page and Midilli models) to 0.1021 (Wang and Singh model) and, among the models with best fits, Midilli and Page result in higher values of R^2 and lower values of χ^2. The Approximation of Diffusion model also stood out with the quality of the fits.

Fits with high R^2 values were also found by Alexandre *et al.* (2013), who studied the drying kinetics of pineapple peels at temperatures from 40 to 60 °C and found that the Henderson and Pabis, Page and Lewis models satisfactorily fitted to the experimental data, but Page model had higher values of R^2 at all temperatures, and by Martins *et al.* (2014), who dehydrated mulungu (*Erythrina velutina*) bark at temperatures of 40, 50, 60 and 70 °C and found that Page model satisfactorily represented the drying process, with R^2 values ≥ 0.99.

Satisfactory fits for the same models tested for citron melon peel flours were detected by Santos et al. (2020), when studying drying kinetics in grape peels at temperatures of 60, 70, 80 and 90 °C, for which Page model was the one that best fitted to the experimental data obtained, and by Barros *et al.* (2020), who fitted mathematical models to the experimental data of the drying kinetics of kino peels and reported that Page and Approximation of Diffusion models were satisfactory.

The parameters 'k' (drying rate constant) and 'n' (internal resistance to drying) of the Midilli model showed variations with the temperatures, with a general trend of increase in 'k' and reduction in 'n'. Martins *et al.* (2015) mentioned that the parameter 'k' can be used as an approximation to characterize the effect of temperature, being directly related to the effective diffusivity in the drying process in the falling rate period, and the liquid diffusivity controls the process. Thus, the higher the magnitude of the parameter 'k', the greater the effective diffusivity in the drying process (Goneli *et al.*, 2017).

In Page model, the values of 'k' and 'n' did not show a defined trend with the variation of drying temperature, with relative variations between temperatures similar to those observed in the parameter's 'k' and 'n' of Midilli model. Santos *et al.* (2017), when drying pitaya (*Hylocereus guatemalensis*) peels, and Martins *et al.* (2014), when dehydrating mulungu barks, reported that the parameter 'k' of all evaluated models (Two Terms, Two-Term Exponential, Henderson and Pabis, Logarithmic, Midilli and Page) increased with the increase in drying temperature, while the parameter 'n' showed no defined trend.

Table 3. Parameters, coefficients of determination (R^2), mean squared deviations (MSD) and chi-squares (χ^2) of mathematical models fitted to the drying curves of citron melon peels at temperatures of 60, 70, 80 and 90 °C

Models		Parameters					
Newton	T (°C)	k			R^2	MSD	χ^2
	60	0.0104			0.9872	0.0422	0.0019
	70	0.0124			0.9902	0.0362	0.0014
	80	0.0124			0.9798	0.0533	0.0030
	90	0.0136			0.9912	0.0335	0.0012
Modified Page	T (°C)	K	n		R^2	MSD	χ2
	60	0.0115	1.0720		0.9933	0.0304	0.0010
	70	0.0135	1.0636		0.9948	0.0264	0.0008
	80	0.0138	1.0852		0.9883	0.0409	0.0018
	90	0.0147	1.0602		0.9956	0.0237	0.0006
Page	T (°C)	k	n		R^2	MSD	χ2
	60	0.0028	1.2931		0.9993	0.0097	0.0001
	70	0.0043	1.2477		0.9994	0.0092	0.0001
	80	0.0023	1.3892		0.9993	0.0102	0.0001
	90	0.0056	1.2092		0.9989	0.0121	0.0002
Henderson and Pabis	T (°C)	a	k		R^2	MSD	χ2
	60	1.0720	0.0115		0.9933	0.0304	0.0010
	70	1.0636	0.0135		0.9948	0.0264	0.0008
	80	1.0852	0.0138		0.9882	0.0409	0.0018
	90	1.0602	0.0147		0.9986	0.0237	0.0006
Two-term exponential	T (°C)	a	k		R^2	MSD	χ2
	60	0.0019	5.4471		0.9869	0.0427	0.0020
	70	0.0022	5.6112		0.9898	0.0368	0.0015
	80	0.0018	6.7623		0.9795	0.0537	0.0032
	90	0.0020	6.6853		0.9909	0.0341	0.0013
Wang and Singh	T (°C)	a	b		R^2	MSD	χ2
	60	-0.0062	0.000008		0.9417	0.0901	0.0088
	70	-0.0070	0.000011		0.9216	0.1021	0.0114
	80	-0.0076	0.000013		0.9519	0.0824	0.0074
	90	-0.0084	0.000016		0.9498	0.0802	0.0071
Logarithmic	T (°C)	a	k	c	R^2	MSD	χ2
	60	1.0942	0.0107	-0.0294	0.9948	0.0270	0.0008
	70	1.0813	0.0127	-0.0243	0.9958	0.0236	0.0006
	80	1.1183	0.0126	-0.0435	0.9910	0.0356	0.0015
	90	1.0796	0.0138	-0.0273	0.9967	0.0204	0.0005
Logarithmic exponential	T (°C)	a	k0	k	R^2	MSD	χ2
	60	0.0873	0.0104	0.1196	0.9872	0.0422	0.0020
	70	0.0969	0.0124	0.1280	0.9902	0.0362	0.0015
	80	0.0706	0.0124	0.1761	0.9798	0.0533	0.0033
	90	0.1031	0.0136	0.1315	0.9912	0.0335	0.0013
Diffusion approximation	T (°C)	a	k	b	R^2	MSD	χ2
	60	-4.1668	0.0223	0.8482	0.9993	0.0101	0.0001
	70	-0.8353	0.0331	0.5594	0.9993	0.0094	0.0001
	80	-38.5963	0.0260	0.9784	0.9989	0.0124	0.0002
	90	-0.2153	0.0730	0.2300	0.9992	0.0104	0.0001

	T (°C)	a	k0	b	k1	R^2	MSD	χ2
Two-terms	60	0.5360	0.0115	0.5360	0.0115	0.9933	0.0304	0.0011
	70	0.5318	0.0135	0.5318	0.0135	0.9948	0.0264	0.0008
	80	0.5424	0.0138	0.5429	0.0138	0.9882	0.0409	0.0020
	90	0.5303	0.0147	0.5299	0.0147	0.9956	0.0237	0.0007
Midilli	T (°C)	a	k	n	b	R^2	MSD	χ2
	60	1.0025	0.0030	1.2851	-0.000003	0.9993	0.0096	0.0001
	70	1.0025	0.0044	1.2404	-0.000002	0.9994	0.0092	0.0001
	80	0.9915	0.0021	1.4104	-0.000009	0.9993	0.0096	0.0001
	90	1.0114	0.0064	1.1808	-0.000009	0.9989	0.0116	0.0002

Figure 3 shows the points representing the drying kinetics data of citron melon peels with fitting curves obtained with Midilli model. There was a more pronounced effect of the difference between the drying temperature of 60 C and the others, with the curves representing the temperatures of 70, 80 and 90 °C occupying positions that were close and little different from each other. The average drying times for temperatures of 60, 70, 80 and 90 °C were 570, 520, 460 and 400 min, respectively.

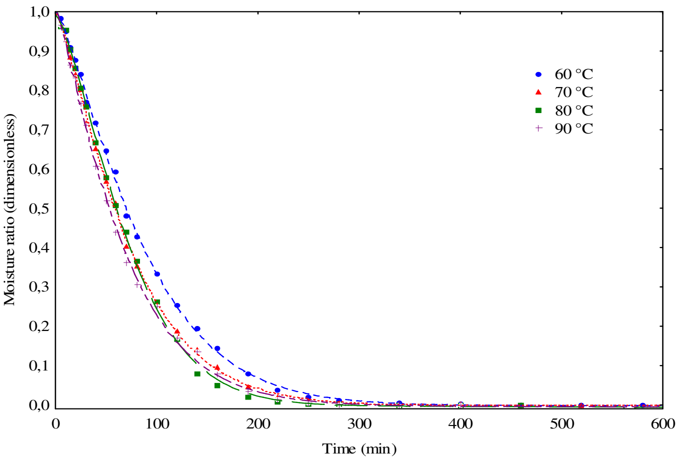

Figure 3. Midilli model fitted to the drying kinetics of citron melon peels at drying temperatures from 60 to 90 °C

The effective diffusivity obtained in the drying of citron melon peels at temperatures from 60 to 90 °C are presented in Table 4. The diffusivity increased consistently with the temperature increments, by more than 30% between 60 and 90 °C. The values obtained, between 1.40×10^{-10} and 1.83×10^{-10} m^2/s, are lower than those determined by Mphahlele *et al.* (2019) for pomegranate peel, with values of 4.05×10^{-10}, 5.06×10^{-10} and 8.10×10^{-10} m^2/s at temperatures of 40, 50 and 60 °C, respectively. Menezes *et al.* (2013) also observed that the effective diffusivity of dehydrated yellow passion fruit bagasse increased from 8.11×10^{-10} to 2.11×10^{-9} m^2/s when the drying temperature increased from 35 °C to 65 °C.

According to Martins *et al.* (2018), diffusivity increases with the increments in temperature due to the increase in the vibration level of water molecules and reduction in water viscosity, which is a measure of the resistance of a fluid to the flow. Variations in this property lead to changes in the diffusion of water in the capillaries of agricultural products, which contributes to a faster diffusion at higher temperatures, in addition to a more intense vibration of water molecules (Martins *et al.*, 2018).

Table 4. Mean values of effective diffusivity (D_{ef}) and coefficients of determination (R^2) obtained in the drying of citron melon peels between 60 and 90 °C

Temperature (°C)	Def (m^2/s)	R^2
60	1.40 × 10-10	0.9355
70	1.67 × 10-10	0.9428
80	1.69 × 10-10	0.9232
90	1.83 × 10-10	0.9426

The values of the effective water diffusivity (D_{ef}) represented in the form of 'ln D_{ef}' as a function of the reciprocal of absolute temperature (1/T) referring to drying of citron melon peels at different temperatures are shown in Figure 4. The increase in diffusivity with the increments in temperature was well described by the Arrhenius equation, with R^2 higher than 0.99, indicating dependence of this property on the drying temperature.

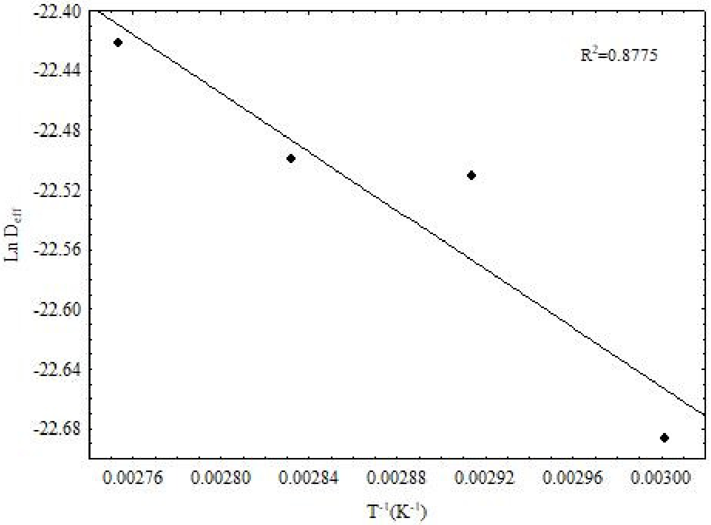

Figure 4. Representation of the Arrhenius equation for effective diffusivity as a function of the inverse of absolute temperature obtained in the drying of citron melon peels

From the values of 'ln D_{ef}' as a function of the inverse of absolute temperature (1/T) (Figure 4), the activation energy (E_a), calculated from the slope of the line, and D_{ef0}, obtained from the linear coefficient of the generated equation, were obtained. This linear regression was used to obtain Equation 22, which represents the effective diffusivity as a function of the drying temperature of the samples.

$$D_{ef} = 2.7767 \times 10^{-9} \exp\left(-\frac{8,18}{T}\right) \qquad (22)$$

According to Equation 22, the activation energy for the diffusion of water in the drying of citron melon peels was 8.18 kJ/mol. Considering that the activation energy refers to the energy needed to break the barrier found by water molecules when they migrate to the surface of the product along the drying process, the higher its value, the lower the diffusivity of water in the product (Silva *et al.*, 2017b). Botelho *et al.* (2018) clarify that the activation energy is the minimum energy required to trigger the diffusive process of water vapor in the product and it can be used to compare drying between different products.

Table 5 shows the mean values of the thermodynamic properties obtained in the drying of citron melon peels between 60 and 90 °C.

Table 5. Mean values of the thermodynamic properties in the drying of citron melon peels at different temperatures

Temperature (°C)	ΔH (kJ/mol)	ΔS (kJ/mol K)	ΔG (kJ/mol)
60	5.4102	-0.4096	141.8790
70	5.3271	-0.4098	145.9765
80	5.2439	-0.4101	150.0765
90	5.1608	-0.4103	154.1789

ΔH - enthalpy; ΔS - entropy; ΔG - Gibbs free energy

Reductions in enthalpy (ΔH) were observed as the drying temperature increased, with values between 5.4102 and 5.1608 kJ/mol, a small but consistent percentage variation for all temperature increments. Enthalpy was positive, a consequence of drying, which requires energy to occur (Santos *et al.,* 2019). Enthalpy is the state function of a chemical reaction that reflects reactions of absorbed or released heat and dissociation of chemical bonds under constant pressure (Kim *et al*., 2010).

Entropy (ΔS), with values between -0.4096 and -0.4103 kJ/mol K, also shows a trend of reduction as the drying temperature increased. Entropy alterations were negative, indicating that diffusion evolved from a disordered initial state, comprising several available sorption sites, to an ordered state at the end of the process, with a greater restriction of molecular movement due to the decrease in sorption sites (Oliveira *et al*., 2020). When $\Delta S < 0$, the process can be characterized as slow, indicating that the material has undergone some kind of physical or chemical aging process, leading it to a state close to its own thermodynamic equilibrium (AnabeL *et al*., 2018).

Gibbs free energy (ΔG), with an increase slightly higher than 8% between temperatures of 60 and 90 °C, showed a behaviour similar to that of enthalpy, with progressive variations at each increment of 10 °C. ΔG increased as the drying temperature increased, a behaviour also evidenced in several agricultural products, for example in the modelling and thermodynamic properties of drying of acuri slices (Santos *et al*., 2019). The positive values indicate that, under these conditions, water diffusion in the product was not spontaneous and that there was consumption of energy from the medium for the reaction to occur (Araujo *et al*., 2017), characterizing the present reaction as endergonic.

According to Oliveira *et al.* (2015), enthalpy changes provide a measure of the energy variation that occurs in the interaction of water molecules with the constituents of the product during sorption processes. Entropy may be associated with the binding or repulsion of water molecules of food components in the system and is associated with spatial arrangement of the water-product relationship. Entropy characterizes, or defines, the degree of order or disorder existing in the water-product system. Gibbs free energy, on the other hand, is an indication of the product's affinity with water, providing a criterion for evaluating water desorption. Changes in Gibbs free energy during the exchange of water between the product and the vicinity are associated with the energy required to transfer water molecules from the vapor state to a solid surface or vice versa.

Conclusions

Citron melon peels subjected to drying are sources of sugars and contain reasonable lipid contents of eleven models fitted to the drying kinetics, at temperatures between 60 and 90 °C, ten resulted in good predictions, especially Midilli, Page and Approximation of Diffusion;

Diffusivity increased consistently with temperature increments, with dependence on temperature well described by the Arrhenius equation;

Drying processes are defined as endergonic, with variations in thermodynamic properties, between temperatures of 60 and 90 °C.

Authors' Contributions

Conceptualization: LSLL, RMFF and AJMQ; Data curation: LSLL; Formal analysis: LSLL, DDFL and WPS; Funding acquisition: RMFF and AJMQ; Investigation: LSLL, DDFL, WPS, CCC and JPS; Methodology: LSLL, RMFF, AJMQ, DDFL and JPS; Project administration and Resources: RMFF and AJMQ; Software: LSLL, RMFF and WPS; Supervision and Validation: RMFF and AJMQ; Visualization and writing-editing: LSLL and DDFL; writing- review and editing: RMFF, AJMQ, WPS, CCC and JPS. All authors read and approved the final manuscript.

Ethical approval (for researches involving animals or humans)

Not applicable.

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Conflict of Interests

The authors declare that there are no conflicts of interest related to this article.

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