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ORIGINAL RESEARCH PAPER

MODELLING OF DRYING KINETICS AND COMPARISON OF TWO PROCESSES: FORCED CONVECTION DRYING AND MICROWAVE DRYING OF CELERY LEAVES (*Apium graveolens* L.)

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The purpose of this work is to compare two processes: forced convection drying and microwave drying of celery leaves (*Apium graveolens* L.). This comparison is based on kinetical parameters, moisture diffusivity, variation of the drying rate and energy consumption calculation of both processes. The drying experiments were carried out at different air temperatures (40-120 °C) and at different microwave powers (100-1000 W).Twenty-two empirical models were used to simulate the thin-layer drying kinetics of celery leaves and the best models were selected using three statistical criteria (R^2, χ^2 and RMSE). Sledz's model proved to be the best for celery leaves drying kinetics description with $0.9962 \leq R^2 \leq 0.9992$, $0.000065 \leq \chi^2 \leq 0.000284$ and $0.007979 \leq \text{RMSE} \leq 0.016683$ for all the studied temperatures and $0.9971 \leq R^2 \leq 0.9989$, $0.000124 \leq \chi^2 \leq 0.000291$ and $0.010910 \leq \text{RMSE} \leq 0.016914$ for all the used powers. Moisture effective diffusivity ranges from 2.22×10^{-12} to 6.40×10^{-11} for convective drying and from 1.18×10^{-11} to 3.13×10^{-10} m^2/s for microwave drying. While in the same order, the activation energies were 36.09 kJ/mol and 77.3 W/g. Regarding the energy consumption, the Specific Electrical Energy Consumption decreased with decreasing temperature or power levels, whereas the opposite was observed with Energy Efficiency. It is clear that many advantages are attributed to microwave drying, including reduced drying time, high drying rate and high moisture diffusivity, low energy consumption and significant drying efficiency, especially when power levels are high.

Keywords: Apium graveolens L., drying kinetics, simulation, moisture diffusivity, activation energy, energy consumption, drying rate

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Introduction

Aromatic plants represent an important resource for human food, due to their valuable properties including medicinal benefits (Soran *et al*., 2014). They find their applications in various fields: food, cosmetic and medical fields. Celery (*Apium graveolens* L.) is one of these plants; it belongs to Umbelliferae family. Endemic to Southern Europe, it is now grown worldwide for human consumption (Kurobayashi *et al.*, 2014). It is a good source of vitamins, minerals and many active ingredients (Madamba and Liboon, 2007), and it is considered among the most desired products due to its taste, texture and low calorie content (González-Buesa *et al.*, 2014). However, one of the main quality attributes of celery is it's crispy nature which depend on degree of lignification of fibrous tissue, dehydration and the degree of alteration of pectic substances (Viña and Chaves, 2003). Therefore, celery is usually dried and used as a soup enhancer because of its characteristic odor, to enhance the taste and aroma of the stew while having therapeutic properties (Demirhan and Özbek, 2011).

Drying is one of the oldest methods of food preservation as well as an important food processing stage (Kayisoglu and Ertekin, 2011). It is also a common industrial preservation method applicable to a wide range of agricultural and industrial products (Zhang *et al.*, 2016). Defined as the removal of moisture from a product by heat, that confer to a product an acceptable moisture level in order to prevent marketing deterioration within a certain period of time, safe storage, processing, or transportation (Nwakuba *et al.*, 2017). It aims to prevent microbial spoilage and chemical alterations thus prolonging shelf-life while realizing space and weight saving (Haksever and Moralar, 2017).

Convective drying is the most universal dehydration method due to its simplicity and easiness of control. However, a long convective process is highly energy consuming, which leads to cell structure collapse during drying and results in inferior quality of dried herbs (Sledz *et al.*, 2017). That is why, the use of microwave rays in the drying of products has become widespread because it minimizes the decline in quality and provides rapid and effective heat distribution in the material as well. Furthermore, high quality product is obtained via microwave drying in addition to the decline in drying period and energy conservation during drying (Alibas and Kacar, 2016).

Given the very large variability and diversity of food and biological products, the best way to characterize the drying behavior of a product is to experimentally measure its drying kinetics (Bonazzi *et al.*, 2008) which are the most important data required for the design and simulation of dryers (Ratti, 2008).

Simulation or mathematical modelling of the drying process and equipment is an important aspect of drying technology in post-harvest processing of agricultural materials (Harish *et al.*, 2014). Numerous models are created to describe the history of the product during the drying process, using parameters such as time, temperature, water content and quality index (Bonazzi and Dumoulin, 2011). The purpose of modelling is to allow the engineers to choose the most appropriate method of drying for a given product as well as to choose suitable operating conditions. Full-scale experimentation for different products and system configurations is sometimes costly and not possible. Therefore, the prediction of drying kinetics of specific crops under various conditions is very useful in the design and optimization of dryers. The use of a simulation model is also a valuable tool for the prediction of drying systems performance (Khazaei and Daneshmandi, 2007).

Previous studies on the modelling of the microwave drying process of celery leaves were reported by Demirhan and Özbek (2011) and Alibas (2014). However, these authors have not studied the information on energy consumption that allows the identification of the best drying process in terms of energy efficiency. Such information will make it easier for the industry to effectively manage drying techniques and thus avoid energy misuse. Moreover, no comparative study between convection drying and microwave drying has been performed on celery leaves, which is the case of the present study whose objectives were: (i) to study the kinetics of celery leaves drying by two methods (forced convection drying and microwave drying), (ii) designate the most appropriate kinetics model, (iii) calculate effective diffusivities, (iv) evaluate the evolution of drying rate kinetics and its variation depending on the moisture content, and finally (v) determine the activation and consumption energies.

Materials and methods

Materials

Fresh celery (*Apium graveolens* L.) was purchased from a local market (Bejaia-Algeria) in November 2016. The leaves were separated from the stems. Before drying experiments, their initial moisture content was determined according to the protocol of Boulekbache-Makhlouf *et al.* (2013) and was 88.16 ±1.14%, their thickness was measured with a Vernier calliper, and the average was found to be 0.4 ±0.07 mm.

Drying equipment and drying procedure

The celery-leaf drying experiments were carried out using two different techniques: hot air drying and microwave drying. Hot air drying (40, 60, 80, 100 and 120 °C) was carried out using a ventilated oven (Memmert, Germany). The microwave drying process (100, 200, 300, 400, 500, 600, 700, 800, 900 and 1000 W) was carried out using a domestic microwave (Maxipower, Model MASMO23S, China). During the drying experiments, 10 g of celery leaves were uniformly spread as a thin layer and the moisture losses were periodically measured using an external analytical accuracy of 0.01 g (RADWAG, WPS 600/C/2, Poland). Three repetitions were performed for each temperature and power, and the data provided were an average of these results. The drying process was applied until the weight of the sample was constant, followed by the determination of the final moisture content of the powders, which is estimated at 7.96 ± 0.11%.

Mathematical modelling of drying curves

Various mathematical models were reported to study the modelling of thin-layer drying kinetics. In this study, twenty-two (22) different expressions were examined (Table 1).

Table 1. Mathematical models given by various authors for drying curves

Name	Mathematical equation	References
Newton	MR=exp(-kt)	Maskan (2000)
Henderson and Pabis	MR=a.exp(-kt)	Nema *et al.* (2013)
Logarithmic	MR=a.exp(-kt)+c	Nema *et al.* (2013)
Page	MR=exp(-kt^n)	Nema *et al.* (2013)
Modified Page 1	MR=exp(-(kt)^n)	Nema *et al.* (2013)
Modified Page 2	MR=exp(-kt)^n	Demirhan and ÖZbek (2009)
Midilli *et al.*	MR=a.exp(-kt^n)+bt	Nema *et al.* (2013)
Two terms	MR=a.exp(-kt)+b.exp(-k_1t)	Nema *et al.* (2013)
Two terms exponential	MR=a.exp(-kt)+(1-a).exp(-kat)	Nema *et al.* (2013)
Approximation of diffusion	MR=a.exp(-kt)+(1-a).exp(-kbt)	Nema *et al.* (2013)
Verma *et al.*	MR=a.exp(-kt)+(1-a).exp(-k_1t)	Nema *et al.* (2013)
Modified Henderson and Pabis	MR=a.exp(-kt)+b.exp(-k_1t)+c.exp(-k_2t)	Vega-Gálvez *et al.* (2014)
Parabolic	MR=a+b.t+c.t^2	Taghian Dinani *et al.* (2014)
Wang and Singh	MR=1+a.t+b.t^2	Nema *et al.* (2013)
Chavez-Mendez *et al.*	MR=(1-(1-L_2)L_1t)$^{1/(1-L_2)}$	Nema *et al.* (2013)
Logistic	MR=b/(1+a.exp(k.t))	Sledz *et al.* (2016)
Sledz *et al.*	MR=b.exp(-kt)/(1+a.exp(k_1.t))	Sledz *et al.* (2016)
Simplified Fick's diffusion equation	MR=a.exp(-k(t/L^2))	Aghbashlo *et al.* (2009)
Weibull	MR=exp(-(t/a)^b)	Aghbashlo *et al.* (2009)
Demir *et al.*	MR=a.exp(-kt)^n+b	Amiri Chayjan and Shadidi (2014)
Taghian Dinani *et al.*	MR=a.exp(-((t-b)/a)^2)	Taghian Dinani *et al.* (2014)
Fernando and Amarasinghe	MR=(1+a.t+b.t^2)/(1+c.t)	Fernando and Amarasinghe (2016)

Note: k, k_1, k_2 – drying coefficients (1/min); *a, b, c,* L_1, L_2 − coefficients of the equations; *n* − exponent; *t*– time (min); *L* – half of thickness (m).

These models estimate that the drying curves respond to an equation of the moisture ratio as a function of drying time. A dimensionless moisture ratio (MR) was calculated using the following equation:

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

where: MR is the moisture ratio, M_t is the moisture content at a specific time t, and M_0 and M_e are the initial and equilibrium moisture content, respectively. However, M_e is relatively small compared with M_t or M_0, hence the error involved in the simplification by assuming that M_e is equal to zero is negligible (H. Darvishi *et al.*, 2016). For this purpose, the moisture ratio can be written in a more simplified form, as follows:

$$MR = \frac{M_t}{M_0} \tag{2}$$

Drying rate

The drying rate (DR) was calculated using equation 3:

$$DR = \frac{M_{t+dt} - M_t}{dt} \tag{3}$$

where DR is the drying rate (kg water/kg dry matter (dm) min), M_{t+dt} and M_t are moisture content at $t+dt$ (kg water/kg dm) and t (kg water/kg dm), respectively, and dt is the time difference (min). The graphical plot of the drying rate with respect to time makes it possible to determine the drying behaviour (Aral and Bese, 2016).

Determination of effective moisture diffusivity

One of the most important parameters in drying kinetics is the effective moisture diffusivity (D_{eff}), which describes the transport of moisture from the material to the environment during the period of falling rates (Aral and Bese, 2016; Motevali *et al.*, 2016; Murthy and Manohar, 2014). For this purpose, the second simplified Fick's equation for diffusion was used to estimate the effective moisture diffusivity (D_{eff}) of celery leaves during their drying, considering constant moisture diffusivity, infinite slab geometry and uniform initial moisture distribution:

$$MR = \frac{8}{\pi^2} \sum_{n=0}^{\infty} \frac{1}{(2n+1)^2} e^{\left(-\frac{(2n+1)^2 \pi^2}{4L^2} D_{eff} t\right)} \tag{4}$$

where D_{eff} is the effective moisture diffusivity (m^2/s), L is the half thickness (drying from both sides) of celery leaves (L=0.0002 m), n is the positive integer and t is the drying time (s).

For a long drying period, the above-mentioned equation can be simplified to the first term of the series and Eq. (4) can be expressed in a logarithmic form as Eq. (5) by taking the natural logarithm of both sides (Madhava Naidu *et al.*, 2016; Zhang *et al.*, 2016):

$$\ln(MR) = \ln\frac{8}{\pi^2} - \frac{\pi^2 D_{eff} t}{4L^2} \tag{5}$$

This is evaluated numerically for the Fourier number, $F_0 = D_{eff} \cdot t/4L^2$ (Hosain Darvishi *et al.*, 2014). To this end, Eq. (5) can be rewritten as:

$$\ln(MR) = \ln\frac{8}{\pi^2} - \pi^2 F_0 \tag{6}$$

Thus:

$$F_0 = -0.101\ln(MR) - 0.0213 \qquad (7)$$

The effective moisture diffusivity was calculated using Eq. (8):

$$D_{eff} = \frac{F_0}{\left(\frac{t}{4L^2}\right)} = \frac{-0.101\ln(MR) - 0.0213}{\left(\frac{t}{4L^2}\right)} \qquad (8)$$

Activation energy

Activation energy is often described using a simple Arrhenius exponential relationship to study the effect of temperature. For this purpose, in the case of hot air drying: equation 9 (Eq. (9)) linking temperature to the effective moisture diffusivity was used (Murthy and Manohar, 2014; Ben Haj Said *et al.*, 2015). However, in the case of microwave drying, the process is not isothermal and temperature cannot be measured (Dadalı *et al.*, 2007; Özbek and Dadali, 2007). Therefore, a modified Arrhenius equation (Eq. (10)), used in previous studies (Hosain Darvishi *et al.*, 2014; Alibas and Kacar, 2016), was applied in this study to calculate the activation energy with STATISTICA software (v.8.0).

$$D_{eff} = D_0 e^{\left(\frac{-E_a}{RT}\right)} \qquad (9)$$

$$D_{eff} = D_0' e^{\left(\frac{-E_a m}{P}\right)} \qquad (10)$$

where D_{eff} is the effective diffusivity (m^2/s), D_0 and D_0' are the pre-exponential factor (m^2/s) in oven and microwave, respectively, R is the universal gas constant (8.314 kJ/mol K), T is the temperature (K), m is the mass of raw sample (g), P is the power (W) and E_a is the activation energy expressed in (kJ/mol) for hot air drying and in (W/g) for microwave drying.

Energy consumption

Energy consumption can be evaluated using two different efficiency indices, namely specific electrical energy consumption (Eq. (11)) or energy efficiency (Eq. (12)) (Tsotsas and Mujumdar, 2014):

$$SEC_e = \frac{3600E}{M_S(X_i - X_f)} \qquad (11)$$

where SEC_e is the Specific Electrical Energy Consumption (MJ/kg water), E is the total electrical energy consumption (kWh), X_i and X_f refer to the initial and final moisture contents (dm), respectively, and M_s is the mass of dry solid matter (in kg).

$$EE = M_s\left(X_i - X_f\right)\frac{\Delta h_v}{3600E}.100 \qquad (12)$$

where *EE* is the Electrical Energy (%) and Δh_v is the evaporation enthalpy of water (2257 kJ/kg, at 100°C).

Statistical analysis

In order to model the experimental data of the drying kinetics, a nonlinear regression analysis was carried out using the STATISTICA software (v.8.0). Three statistical parameters, i.e., determination coefficient (R^2), chi-square (χ^2) and root mean square error (RMSE), were used to determine the ability of the tested mathematical models to represent the experimental data. The best model had the highest R^2 value and the lowest χ^2 and RMSE values (Chayjan *et al.*, 2013). These parameters were calculated as follows:

$$\chi^2 = \frac{\sum_{i=1}^{N}(MR_{pre,i} - MR_{\exp,i})^2}{N - z} \tag{13}$$

$$RMSE = \sqrt{\frac{\sum_{i=1}^{N}(MR_{pre,i} - MR_{\exp,i})^2}{N}} \tag{14}$$

where $MR_{exp,i}$ is the experimental moisture ratio, $MR_{pre,i}$ is the predicted moisture ratio, N is the number of observations and z is the number of constant parameters in the model equation.

Concerning the differences between D_{eff}, SEC_e and *EE* values, they were tested by the univariate ANOVA test. The significance of the differences between these values was determined by Tukey test at $P \leq 0.05$. The calculations were performed using Minitab 17 software.

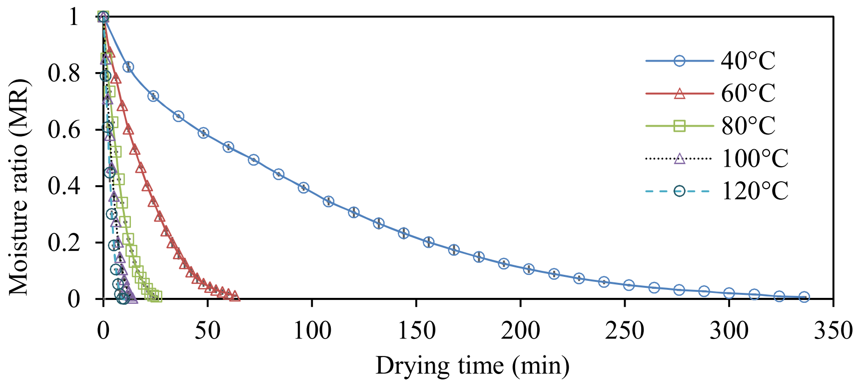
Results and discussion

Drying curves

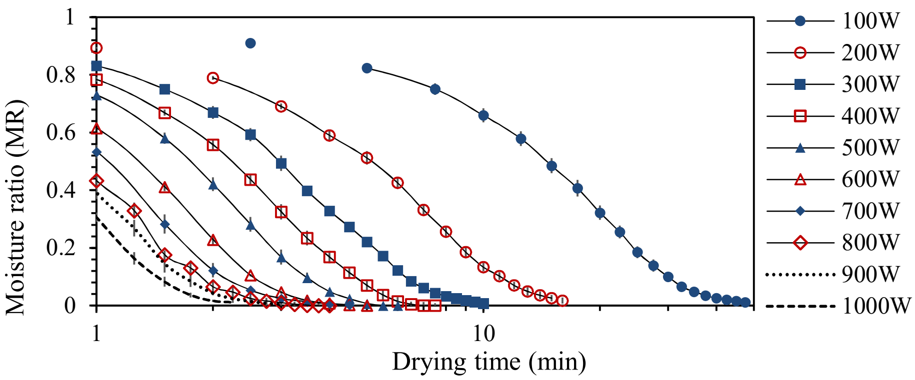
The variation of the moisture content as a function of the celery-leaf drying time at different temperatures (40-120 °C) and at different microwave powers (100-1000 W) are shown in Figure 1 (a) and (b), respectively.

As shown in figure 1 (a) and (b), the drying process is characterized by a gradual decrease in the moisture content as drying time evolves. This decrease is fast at the beginning of the drying process and then slows down gradually as the drying process progresses. This can be explained by the fact that during the first phase of the drying process, the water content in the matrix is high, which leads to a large diffusion of moisture from the product (Alibas and Kacar, 2016). However, at an advanced drying process stage, we observed a reduction of the energy absorbed by the product surface. This is due to the lower water content of the partially dried product and to the dry surface, which prevents the penetration of heat and the migration of water (Ben Haj Said *et al.*, 2015).

It is also noted that temperature and microwave power levels have a significant effect on the moisture content. Indeed, it is clear from the curves that the decrease in the moisture content becomes faster with the increase of both temperature and power levels. Thus, the drying time required to achieve the lowest moisture content was reduced from 338 to 10.83 min for temperatures ranging from 40 to 120 °C and from 48.5 to 2.66 min for powers ranging from 100 to 1000 W. These results have shown that an increase of temperature or power levels leads to a reduction of drying time, which are in accordance with reported observations on convective drying (Faal *et al.*, 2015; İZLİ, 2017; Nguyen *et al.*, 2015; Xiao *et al.*, 2015) as well as those on microwave drying (Amiri Chayjan *et al.*, 2015; H. Darvishi *et al.*, 2016) of different food products other than celery. When comparing processes in terms of reduced drying time, a significant reduction has been observed when applying the microwave drying process, especially when applying high power levels; this could be explained by the fact that one of the advantages of microwave drying is a short processing time (Amiri Chayjan *et al.*, 2015).



(a)



(b)

Figure 1. Moisture ratio profiles of celery leaves during convective drying at various temperatures (a) and microwave drying at various power levels (b).

Modelling of the experimental data

The modelling of drying kinetics allows predicting the drying kinetics of many agricultural and food materials and to evaluate the end point of the process. Several models were reported in the literature. However, in recent years new models have been proposed by several authors, such as the models proposed by Taghian Dinani *et al.* (2014), Fernando and Amarasinghe (2016) and Sledz *et al.* (2016). To better estimate the model that interprets the kinetics of drying celery leaves, twenty-two (22) models (Table 1) were tested and the results were evaluated on the basis of the results of the following statistical parameters: coefficient of determination (R^2), chi-square (χ^2) and root mean square error (RMSE). Of the 22 models tested (Table 1), five models describe the drying behavior of celery leaves in the case of forced convection process (Table 2), and four models describe their behavior in the case of microwave drying (Table 3). Indeed, all these models confirmed the goodness of fit with the R^2 values closest to unity and the lowest χ^2 and RMSE values (Table 2 and 3). However, among these models, the highest R^2 values and the lowest χ^2 and RMSE values were obtained with the Sledz *et al.* (2016) model with $0.9962 \leq R^2 \leq 0.9992$, $0.000065 \leq \chi^2 \leq 0.000284$ and $0.007979 \leq$ RMSE $\leq$0, 016683 and $0.9971 \leq R^2 \leq 0.9989$, $0.000124 \leq \chi^2 \leq 0.000291$ and $0.010910 \leq$ RMSE≤ 0.016914 for both forced convection and microwave drying, respectively. Consequently, this model was chosen as the optimal model for its satisfactory description of the drying characteristics of the celery leaves. This model was also chosen in previous studies to describe the drying behaviour of parsley leaves (Sledz *et al.*, 2016) and basil leaves (Sledz *et al.*, 2017).

Effective moisture diffusivity

The effective moisture diffusivities (D_{eff}) were estimated using Eq. (8); the values are summarized in Table 4. These values show a variation ranging from 2.22×10^{-12} to 6.40×10^{-11} m^2/s and from 1.18×10^{-11} to 3.13×10^{-10} m^2/s for forced convection and microwave drying, respectively, shifting from the lowest temperatures and power levels to the highest ones. This indicates that, the higher the temperature or power levels, the higher D_{eff} values. In the case of convective drying, the increase in D_{eff} values in parallel with high temperature levels can be explained by the fact that the rise in temperature strongly activates water molecules, accelerating their transfer to the surface of the matrix (Amami *et al.*, 2017; Aral and Bese, 2016). However, its increase in the case of microwave drying, is related to the energy absorbed by moisture, which increases with the increment of microwave power level (Zhu *et al.*, 2015).

The D_{eff} values obtained in this study fall within the general range defined by Azeez *et al.* (2017), which is 10^{-8} to 10^{-12} m^2/s for food products. These values are also included in the range of values obtained for food sheets in other studies: for example, 2.55×10^{-12} to 8.83×10^{-12} m^2/s for *Allium roseum* leaves at temperatures ranging from 40 to 60 °C (Ben Haj Said *et al.*, 2015), 0.2×10^{-12} to 9.4×10^{-12} m^2/s for rosemary leaves at temperatures ranging from 40 to 60 °C (Bensebia and Allia, 2015), 3.982×10^{-11} to 2.073×10^{-10} m^2/s for the drying of mint leaves at power levels ranging from 180 to 900 W (Özbek and Dadali, 2007), 2.168×10^{-10} to 7.899×10^{-10} m^2/s for drying basil leaves at power levels ranging from 180 to 900W (Demirhan and Özbek, 2009), 6.3×10^{-11} to 2.19×10^{-10} m^2/s for the drying of coriander leaves in power levels ranging from 180 to 360W (Sarimeseli, 2011), 2.61×10^{-11} to 9.24×10^{-11} m^2/s for the drying of kaffir lime leaves (Tasirin *et al.*, 2014).

Table 2. The values of the drying constants and coefficients and the statistical parameters of the best models for all the drying temperatures.

Models	T(°C)	The drying constants and coefficients				Statistical parameters		
		k	**a**	**b**	**n**	**R^2**	**χ^2**	**RMSE**
Midilli *et al.*	40°C	0.008632	0.922385	-0.000159	1.006303	0.9954	0.000292	0.017025
	60°C	0.022569	0.959861	-0.000602	1.195996	0.9976	0.000195	0.013898
	80°C	0.067321	0.963927	-0.000908	1.233270	0.9992	0.000063	0.007898
	100°C	0.121443	0.971145	-0.001835	1.290154	0.9979	0.000186	0.013528
	120°C	0.161294	0.952268	-0.001938	1.419678	0.9960	0.000366	0.018935
Logistic		**k**	**a**	**b**	**n**	**R^2**	**χ^2**	**RMSE**
	40°C	0.013100	1.145222	1.913492		0.9941	0.000378	0.019379
	60°C	0.074171	0.615314	1.545546		0.9969	0.000250	0.015749
	80°C	0.192628	0.655043	1.606302		0.9989	0.000086	0.009224
	100°C	0.366076	0.492141	1.459457		0.9974	0.000224	0.014894
	120°C	0.586112	0.329364	1.281450		0.9956	0.000399	0.019813
Sledz *et al.*		**k**	**a**	**b**	**k_1**	**R^2**	**χ^2**	**RMSE**
	40°C	0.008567	0.008489	0.922045	0.019669	0.9962	0.000243	0.015537
	60°C	0.036973	0.021661	1.009751	0.094452	0.9984	0.000125	0.011136
	80°C	0.066750	0.172515	1.158395	0.175024	0.9992	0.000065	0.007979
	100°C	0.133254	0.081500	1.085989	0.371006	0.9980	0.000171	0.012989
	120°C	0.204981	0.032171	1.035115	0.686821	0.9969	0.000284	0.016683
Taghian Dinani *et al.*		**a**	**b**	**c**		**R^2**	**χ^2**	**RMSE**
	40°C	2.580211	-277.943	270.6528		0.9952	0.000309	0.017532
	60°C	1.559617	-30.3095	43.84813		0.9978	0.000164	0.012744
	80°C	1.691476	-12.6811	17.19516		0.9993	0.000060	0.007729
	100°C	1.406182	-5.18789	8.749976		0.9980	0.000175	0.013159
	120°C	1.168085	-2.27135	5.339614		0.9964	0.000322	0.017794
Fernando and Amarasinghe		**a**	**b**	**c**		**R^2**	**χ^2**	**RMSE**
	40°C	-0.003633	0.000002	0.008343		0.9925	0.000481	0.021867
	60°C	-0.030337	0.000234	0.008342		0.9982	0.000147	0.012080
	80°C	-0.079351	0.001610	0.021501		0.9990	0.000097	0.009811
	100°C	-0.148013	0.005543	0.015990		0.9977	0.000197	0.013966
	120°C	-0.217602	0.011769	0.008900		0.9963	0.000330	0.018034

Statistical analysis of the D_{eff} values obtained for the same drying process showed that there were no significant differences between the values obtained for 40 and 60 °C in the case of oven drying and that above 60 °C, significant differences were observed from one temperature level to another. In the case of microwave drying, the application of low power levels (100-200 W) did not produce significant differences within neither the obtained D_{eff} values, nor for powers ranging from 300 to 500 W. However, when applying powers greater than or equal to 600 W, significant differences within D_{eff} values are observed, although overlaps have been observed for some values (800-900 W). Statistical analysis of D_{eff} values for both drying processes showed that the values obtained at 40, 60 and 80°C are not statistically different from those obtained at 100 and 200 W. The value obtained at 100 °C is not statistically different from those obtained at 200 and 300 W and the value obtained at 120 °C is not statistically different from those obtained at 300, 400 and 500 W. However, above 500 W, the D_{eff} values are higher and are not comparable to the D_{eff} values of the hot air drying.

Table 3. The values of the drying constants and coefficients, and statistical parameters of the best models for all microwave power levels.

Models	P(W)	The drying constants and coefficients				Statistical parameters		
		k	**a**	**b**	n	**R^2**	**χ^2**	**RMSE**
Midilli *et al.*	100W	0.007540	0.943359	-0.000117	1.660750	0.9973	0.000267	0.016231
	200W	0.053907	0.940993	-0.000810	1.525211	0.9978	0.000198	0.014011
	300W	0.092051	0.916526	-0.000143	1.720028	0.9978	0.000207	0.014305
	400W	0.148047	0.920624	-0.002425	1.727705	0.9966	0.000337	0.018235
	500W	0.210106	0.913292	-0.001894	1.827755	0.9962	0.000385	0.019443
	600W	0.388437	0.913989	-0.000412	1.774091	0.9967	0.000315	0.017552
	700W	0.516362	0.919773	0.000101	1.790402	0.9967	0.000317	0.017593
	800W	0.752570	0.935550	0.000314	1.734070	0.9964	0.000342	0.018239
	900W	0.877581	0.953976	0.000752	1.796015	0.9974	0.000273	0.016241
	1000W	1.141968	0.954619	0.000184	1.826829	0.9981	0.000209	0.014170
Logistic		**k**	**a**	**b**		**R^2**	**χ^2**	**RMSE**
	100W	0.131605	0.191865	1.148881		0.9977	0.000224	0.014883
	200W	0.336599	0.259434	1.206440		0.9981	0.000179	0.013331
	300W	0.644602	0.171465	1.099195		0.9982	0.000169	0.012932
	400W	0.910084	0.144493	1.071069		0.9964	0.000360	0.018881
	500W	1.227753	0.121457	1.044074		0.9963	0.000377	0.019282
	600W	1.576569	0.150404	1.076553		0.9973	0.000260	0.015991
	700W	1.866704	0.147288	1.081388		0.9973	0.000256	0.015846
	800W	2.196001	0.168872	1.120602		0.9970	0.000278	0.016502
	900W	2.506359	0.147411	1.122133		0.9978	0.000222	0.014715
	1000W	2.983243	0.134449	1.108976		0.9985	0.000163	0.012573
Sledz *et al.*		**k**	**a**	**b**	**k_1**	**R^2**	**χ^2**	**RMSE**
	100 W	0.023217	0.064906	1.054098	0.139532	0.9983	0.000171	0.012979
	200W	0.065778	0.093075	1.070496	0.345386	0.9985	0.000140	0.011784
	300W	0.066451	0.098644	1.048313	0.653872	0.9983	0.000153	0.012309
	400W	0.170734	0.033976	1.004625	1.026550	0.9974	0.000263	0.016093
	500W	0.197903	0.033308	0.997699	1.372267	0.9971	0.000291	0.016914
	600W	0.272456	0.041468	1.003781	1.749965	0.9980	0.000182	0.013359
	700W	0.280762	0.052857	1.021320	1.999943	0.9978	0.000212	0.014367
	800W	0.350018	0.061307	1.045710	2.331453	0.9975	0.000237	0.015189
	900W	0.352181	0.057968	1.062325	2.661316	0.9982	0.000182	0.013264
	1000W	0.374240	0.057594	1.060134	3.153385	0.9989	0.000124	0.010910

		a	b	c		R^2	χ^2	RMSE
Taghian Dinani *et al.*	100 W	1.019134	-5.33801	23.45080		0.9979	0.000208	0.014345
	200W	1.070888	-2.96095	9.143665		0.9983	0.000154	0.012369
	300W	0.972649	-0.901775	4.782168		0.9982	0.000173	0.013106
	400W	0.956344	-0.502690	3.421203		0.9968	0.000317	0.017719
	500W	0.937920	-0.260336	2.556637		0.9964	0.000360	0.018851
	600W	0.964158	-0.326289	1.982138		0.9973	0.000256	0.015861
	700W	0.967234	-0.261344	1.672520		0.9972	0.000268	0.016231
	800W	1.001324	-0.280401	1.418352		0.9969	0.000289	0.016835
	900W	1.001122	-0.188802	1.241097		0.9977	0.000231	0.015021
	1000W	a=0.990658	-0.130269	1.044537		0.9984	0.000536	0.022083

Table 4. Average effective diffusivity for celery leaves at different temperatures and at different microwave powers

Temperatures	Diffusivities (m^2/s)	Powers	Diffusivities (m^2/s)
40°C	$2.22\times10^{-12}\pm 1.22\times10^{-13dH}$	**100W**	$1.18\times10^{-11}\pm 3.48\times10^{-13fH}$
		200W	$2.30\times10^{-11}\pm 2.39\times10^{-12fGH}$
60°C	$9.30\times10^{-12}\pm 7.96\times10^{-13dH}$	**300W**	$5.91\times10^{-11}\pm 3.09\times10^{-12eEF}$
		400W	$7.65\times10^{-11}\pm 8.81\times10^{-12eE}$
80°C	$2.53\times10^{-11}\pm 2.04\times10^{-12cGH}$	**500W**	$7.91\times10^{-11}\pm 1.86\times10^{-11eE}$
		600W	$1.70\times10^{-10}\pm 1.24\times10^{-11dD}$
100°C	$4.44\times10^{-11}\pm 9.05\times10^{-13bFG}$	**700W**	$2.12\times10^{-10}\pm 3.05\times10^{-12cC}$
		800W	$2.51\times10^{-10}\pm 3.38\times10^{-12bB}$
120°C	$6.40\times10^{-11}\pm 9.12\times10^{-12aEF}$	**900W**	$2.59\times10^{-10}\pm 1.62\times10^{+11bB}$
		1000W	$3.13\times10^{-10}\pm 9.33\times10^{-12aA}$

Table shows mean values with standard deviation of the mean.

^{a,b} - same index letters indicate that the mean values are not significantly different at a confidence level of 95 % ($p \leq 0.05$)for the same process.

^{A,B} - same index letters indicate that the mean values are not significantly different at a confidence level of 95 % ($P \leq 0.05$) between the two process.

Drying rate versus drying time

The drying rates were calculated using Eq. (3) and their variations as a function of the drying time at different temperatures and power levels are shown in Figure 2 (a) and (b), respectively.

According to figure 2 (a) and (b), the drying process of the studied samples occurs through two steps: a rapid heating period and a falling rate period. The rapid drying period has a variable duration depending on the process as well as the applied temperature or power levels. By moving from low to high levels of drying, the duration of the rapid drying period varied from 8 to 0.66 min (convective drying) and from 2 to 1 min (microwave drying). As for the falling period, it begins just after the completion of the rapid drying period and continues until the end of the drying process. It was during the latter that the drying took place. The mechanism involved in these processes is diffusion, which influences the elimination of water. Higher drying rates were observed at the beginning of the process, and then decreased gradually. This can be explained by the fact that, initially, the higher the temperature or the microwave radiation energy is absorbed by the water on the product surface, the more rapidly the drying occurs. However, with the progress in drying time, the product becomes dry, which hinders the penetration of heat into the matrix, leading to the decrease of the drying rate. The same trend was observed with all temperatures and power levels. However, the latter had an important effect on both the drying rate and the drying time. Indeed, when the temperature or the applied power increased, the drying rate increased and the drying time decreased in parallel. Similar results have been reported in the literature in previous studies conducted by Minaei *et al.* (2011), Hosain Darvishi *et al.* (2014), Zhu *et al.* (2015) and Motevali *et al.* (2016) on microwave drying and those conducted by Murthy and Manohar (2014) and Aral and Bese (2016), who investigated convection drying characteristics.

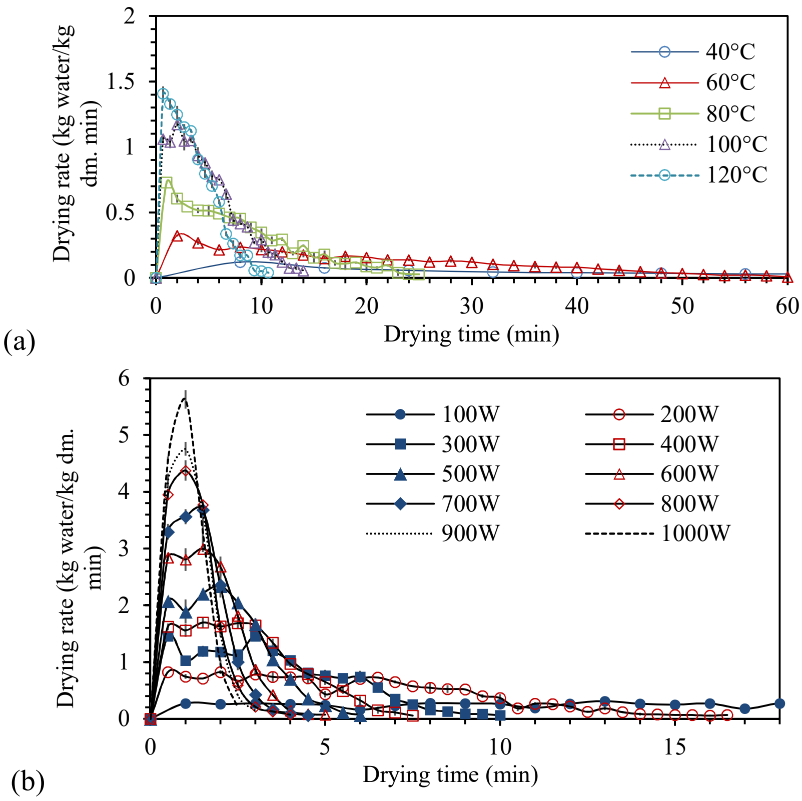
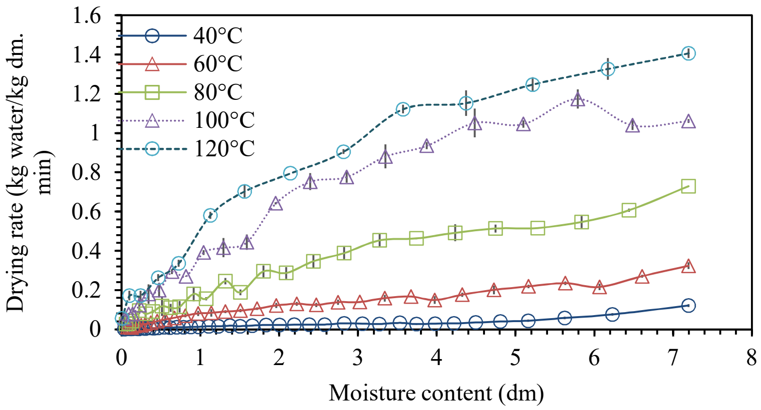


Figure 2. Variations of drying rate as function of drying time at different temperatures of hot air drying (a) and at different microwave powers (b).

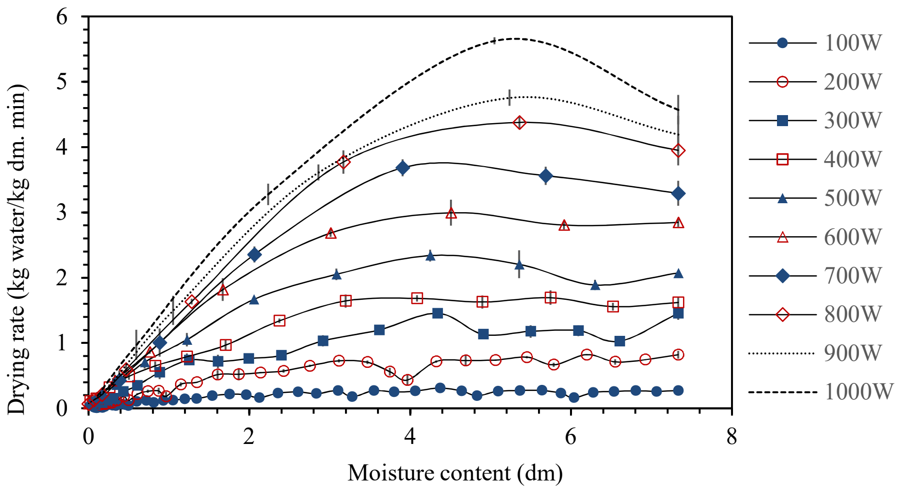
The analysis of the results shows that high drying rates were obtained during the application of the microwave process compared to the drying rates obtained by hot air drying. This is explained by the fact that microwaves have the advantage of reducing the drying time due to the rapid absorption of energy by water molecules, thus causing rapid evaporation of water, resulting in high drying rates (Doymaz *et al.*, 2016).

Drying rate versus moisture content

The drying rate variations with respect to the moisture content at different drying temperatures and microwave power levels are represented in Figure 3 (a) and (b), respectively.



(a)



(b)

Figure 3. Variations of drying rate as function of moisture content at different temperatures of hot air drying (a) and at different microwave powers (b)

Figure 3 (a) and (b) show a proportional relationship between the drying rate and the moisture content. However, differences in appearance are observed according to the drying process applied as well as for the different microwave powers. In fact, as can be observed in the case of convection drying, the highest drying rates are observed when the moisture content of the sample is highest then decrease with the decrease of the moisture content of the sample.

In the case of the microwave, periods of constant drying rates have been observed under particularly low microwave power drying conditions followed by a decrease as the moisture content of the sample drops. Whereas for the power values particularly of high frequency, the periods of constant drying rate have been replaced by an increase in drying rates which are in turn followed by a decrease.

These figures also show the influence of temperature or power level on the drying rate. Indeed, regardless of the water content in the sample, the highest drying rates are induced when the temperature or power level is increased, so that, at a temperature of 40 °C, the average drying rate was about 0.02 kg water/kg dm min. This increases with the application of higher temperatures to achieve an average drying rate of 0.65 kg water/kg dm min at 120 °C. For microwave drying, the average drying rates evolved from 0.15 kg water/kg dm min at the lowest power level (100 W) to 2.93 kg water/kg dm min at the highest level (1000 W). The same rates observed in this study were observed by many authors, such as Khazaei and Daneshmandi (2007) and Kumar *et al.* (2012), who studied the effect of temperature and Dadalı *et al.* (2007) and Alibas and Kacar (2016) who studied microwave drying.

Activation energy

The activation energies of both drying processes: convection and microwave were determined by estimating the coefficient values of equations (9) and (10). The values obtained were estimated at 36.09 kJ/mol and 77.3 W/g for both drying processes: convection and microwave, respectively.

The E_a values obtained when convective drying fall within the general range defined previously (Aral and Bese, 2016; Kaveh and Amiri Chayjan, 2017), which is 12.7 to 110 kJ/mol, determined for fruit and vegetables, and they are comparable to values reported in the literature, although the drying conditions are different. They are all particularly comparable to values in the range of 31.28 and 35.23 kJ/mol for the drying of nectarine slices at temperatures ranging from 50 to 70 °C (Alaei and Chayjan, 2015) and values of 31.94 and 34.49 kJ/mol for seeds of squash at temperatures of 50 and 80 °C, respectively (Chayjan *et al.*, 2013), and those of melon sheaths varying between 27.6 and 45.3 kJ/mol (Golpour *et al.*, 2016). These values are also similar to those (31.19 kJ/mol) obtained during the drying of tomato slices at 50-70 °C (Azeez *et al.*, 2017), mango ginger (32.6 kJ/mol) at 40-70 °C (Murthy and Manohar, 2014), unripe Cardaba banana (38.46 kJ/mol) at 50-70 °C (Olawoye *et al.*, 2017), potato variety *Golden delicious* (35.3 kJ/mol) at 30-60 °C (Cruz *et al.*, 2014) and pumpkin (33.74 kJ/mol) at 30-70 °C (Guiné *et al.*, 2011).

On the other hand, the E_a values obtained during microwave drying are different from those reported in the literature for the drying of many products, such as the values ranging from 17.96 to 21.38 W/g for kiwi slices (H. Darvishi *et al.*, 2016) and 28.68087 W/g for *Hypericum perforatum L.* (Alibas and Kacar, 2016), 5.54 W/g for okra (Dadalı *et al.*, 2007), 14.67 W/g for pepper (Hosain Darvishi *et al.*, 2014), 11.41 W/g for basil leaves (Demirhan and ÖZbek, 2009) and 24.7 W/g for the yam of elephant foot yam (Harish *et al.*, 2014). However, these values are particularly comparable to the E_a reported by Zhu *et al.* (2015), which is about 77.0485 W/g for Ximeng lignite.

Energy consumption

The calculation results for SEC_e and EE are illustrated in Table 5.

Table 5. Result of SEC_e and EE calculations

T(°C)	Convection drying		P(W)	Microwave drying	
	SEC_e (MJ/kg H_2O)	***EE* (%)**		**SEC_e (MJ/kg H_2O)**	***EE* (%)**
40°C	$2.61\times10^{+09}\pm$ $9.11\times10^{+07aA}$	$9.13\times10^{-05}\pm$ 3.22×10^{-06eL}	**100W**	$3.00\times10^{+08}\pm$ $1.00\times10^{+07aC}$	$7.53\times10^{-04}\pm$ 2.51×10^{-05jJK}
			200W	$1.02\times10^{+08}\pm$ $6.42\times10^{+06bEF}$	$2.21\times10^{-03}\pm$ 1.38×10^{-04iI}
60°C	$4.88\times10^{+08}\pm$ $1.25\times10^{+07bB}$	$4.88\times10^{-04}\pm$ 1.23×10^{-05dKL}	**300W**	$6.47\times10^{+07}\pm$ $8.87\times10^{+05cEFGH}$	$3.49\times10^{-03}\pm$ 4.79×10^{-05hH}
			400W	$5.24\times10^{+07}\pm$ $2.66\times10^{+06dEFGH}$	$4.31\times10^{-03}\pm$ 2.18×10^{-04gG}
80°C	$1.95\times10^{+08}\pm$ $6.64\times10^{+06cD}$	$1.22\times10^{-03}\pm$ 4.18×10^{-05cJ}	**500W**	$4.16\times10^{+07}\pm$ $1.42\times10^{+06deFGH}$	$5.43\times10^{-03}\pm$ 1.81×10^{-04fF}
			600W	$3.56\times10^{+07}\pm$ $1.52\times10^{+06efEFGH}$	$6.34\times10^{-03}\pm$ 2.68×10^{-04eE}
100°C	$1.15\times10^{+08}\pm$ $6.01\times10^{+06cE}$	$2.06\times10^{-03}\pm$ 1.04×10^{-04bI}	**700W**	$3.10\times10^{+07}\pm$ $8.81\times10^{+06efgFGH}$	$7.29\times10^{-03}\pm$ 2.11×10^{-04dD}
			800W	$2.76\times10^{+07}\pm$ $1.05\times10^{+06fghGH}$	$8.19\times10^{-03}\pm$ 3.05×10^{-04cC}
120°C	$9.27\times10^{+07}\pm$ $2.27\times10^{+06cEFG}$	$2.57\times10^{-03}\pm$ 6.23×10^{-05aI}	**900W**	$2.25\times10^{+07}\pm$ $1.06\times10^{+06ghGH}$	$1.01\times10^{-02}\pm$ 4.74×10^{-04bB}
			1000W	$1.72\times10^{+07}\pm$ $2.85\times10^{+06hH}$	$1.31\times10^{-02}\pm$ 2.15×10^{-04aA}

Table shows mean values with standard deviation of the mean.

[a,b] - same index letters indicate that the mean values are not significantly different at a confidence level of 95 % ($p \leq 0.05$)for the same process.

[A,B] - same index letters indicate that the mean values are not significantly different at a confidence level of 95 % ($p \leq 0.05$) between the two process.

As it can be observed, SEC_e decreases when moving from a low temperature or power level to higher temperatures or power levels for convection and microwave drying, respectively. Indeed, as indicated in Table 5, SEC_e ranges from $9.29\times10^{+7}$ to $2.61\times10^{+9}$ MJ/kg in the case of convective drying and from $2.24\times10^{+7}$ to $3.00\times10^{+8}$ MJ/kg for microwave drying. The statistical analysis showed significant differences between SEC_e values obtained at 40, 60 and 80 °C. In the case of microwave drying, the use of low power levels (100-400 W) produced significant differences within the obtained SEC_e values. When applying powers greater than or equal to 400 W, with significant differences between SEC_e values, overlaps were observed for some values. It should be noted that the decline in SEC_e values has been reported by many authors, such as Amiri Chayjan and Shadidi (2014), who found in their study on bean drying, that SEC_e values ranged from $13.36\times10^{+6}$ to $64.30\times10^{+6}$ kJ/kg for the temperature range of 40 to 70°C. Chayjan *et al.* (2013) reported in their study on squash seed values ranging from $2.303\times10^{+6}$ to $0.783\times10^{+6}$ kJ/kg for 50-80 °C and H. Darvishi *et al.* (2016) reported values of 7.79 - 16.20 MJ/kg for kiwi slices drying.

Concerning *EE* variations (Table 5), an opposite behavior to SEC_e was observed, i.e., *EE* values increased when moving from low temperatures or power levels to high temperatures or power levels. The values obtained for this parameter range from 9.13×10^{-5} to 2.57×10^{-3}, from 40 to 120 °C, and from 7.53×10^{-4} to 1.31×10^{-2} for power levels ranging from 100 to 1000 W. The statistical analysis of these results showed significant differences when moving from one level to another for both temperatures and power levels. It should be noted that the opposite trends for SEC_e and *EE* were also observed by Tsotsas and Mujumdar (2014).

Statistical analysis of the SEC_e values obtained for both drying processes showed that the most important SEC_e values are obtained at 40, 60, 80 °C and 100 W which are statistically different. The SEC_e values of the other temperatures (100 and 120 °C) and powers between 200 and 700 W do not show any statistical difference, but they are different from those obtained at powers between 800 and 1000 W, which in turn do not show a statistical difference from the SEC_e values of powers between 300 and 700 W.

The statistical analysis of the *EE* values obtained for both drying processes showed that the lowest values are induced at low temperatures (40 and 60 °C) which are not statistically different from each other and the highest values are obtained at powers ranging from 300 to 1000 W and which are also statistically different from each other. However, the absence of significant differences in *EE* values is observed at temperatures of 60 to 120 °C overlapping the *EE* values obtained at 100 and 200 W.

By comparing both drying processes it appears that in general, the energy consumption (most important SEC_e value) was obtained for the convection drying and the energy efficiency (most important *EE* value) was obtained for microwave drying. This suggests that the energy consumption is proportionally related to the drying time while the efficiency is inversely related to it.

Conclusions

The characteristics of thin-layer drying kinetics of celery leaves were studied by two methods: convection and microwave drying. In order to simulate the experimental data of the drying kinetics, twenty-two (22) mathematical models were tested and the results allowed choosing Sledz *et al.* model as the best model to describe the drying behaviour of celery leaves for both methods reinforced by the highest R^2 values and the lowest $\chi 2$ and RMSE values. A comparison between both methods was made on the basis of drying parameters such as drying time, variation in drying rate as a function of time and as a function of moisture content, calculation of diffusivities and energy consumption. The results concluded that increasing the drying temperature or microwave power level resulted in a reduction in drying time and that this reduction is more significant when applying microwave drying.

As far as the effective moisture diffusivity is concerned, it rises with increasing temperature or power levels and the most important diffusivities are induced when applying high levels of microwave power. The variation in drying rate with time and its variation with moisture content increases with increasing temperature or power level and the highest drying rates are also obtained with high levels of microwave power. With regard to energy consumption and drying efficiency, calculations have shown that energy consumption is low and drying efficiency is high when applying high convection temperatures or microwave power levels. The results have also shown that energy consumption is higher when applying oven drying while drying efficiency is obtained for the microwave drying process.

Based on the obtained results, it can be concluded that the microwave drying technique was better for celery leaves drying, given the many advantages reported in this study, including reduced drying time, high drying rates, high diffusivities, low energy consumption and drying efficiency.

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