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ORIGINAL PAPER

Moisture Diffusivity Coefficient and Convective Drying Modelling of Murta (*Ugni molinae* Turcz): Influence of Temperature and Vacuum on Drying Kinetics

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Abstract In this study, murta (*Ugni molinae* Turcz) or murtilla berries were dried in single layer at air temperatures of 50, 60 and 70 °C under vacuum and atmospheric pressure conditions. The effect of drying air temperature and vacuum on the basic dehydration characteristics of murta was determined. For the kinetic modelling, ten mathematical expressions were fitted to the experimental data. Kinetic parameters and diffusion coefficients as evaluated by an Arrhenius-type equation, showed temperature dependency. Fick's second law was used to calculate the effective moisture diffusivity that varied from 3.10 to 11.27×10^{-10} m^2/s and from 5.50 to 11.30×10^{-10} m^2/s with activation energy values of 59.27 and 34.30 kJ/mol for atmospheric pressure and vacuum drying, respectively. According to the statistical tests applied, the Midilli–Ku uk model obtained the best-fit quality on experimental data, followed closely by the Weibull distribution model, the Page and the modified Page models.

Keywords Murta berries · Kinetic modelling · Vacuum drying · Diffusion coefficient · Weibull distribution

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Introduction

Murta (*Ugni molinae* Turcz), also known as murtilla or Chilean guava, is an edible berry from a forest understorey shrub growing wildly in the southern regions of Chile. It belongs to the Myrtaceae family and is found mainly in woodland edges of the coastal mountains of the Andes (Hoffmann 1991; Pastenes et al. 2003). The characteristic aroma of murta berries may be described as a mixture of fruity, sweet and floral notes, due to volatile compounds similar to those found in many aromatic tropical and traditional fruits widely consumed around the world (Scheuermann et al. 2008). In its growing locations, murta is highly appreciated and is consumed as a fresh fruit during the summer, but is also processed to jam, preserve cooked with quince in syrup, confections, juice and liquor. Murta and other berries from South America possess a rich and diversified composition of bioactive compounds with health-promoting properties that have been intensively studied (Speisky et al. 2008; Ruiz et al. 2010; Schreckinger et al. 2010). Its economical importance is gaining ground and serious efforts to domesticate the wild fruits have been undertaken and resulted in two new varieties; "South Pearl-

INIA" was registered by Seguel and Montenegro (2008) and "Red Pearl-INIA" by Seguel and Montenegro (2010).

In general, the consumption of berries has increased during the last decade and berries of different kinds are widely consumed in many countries (Heinonen 2007; Szajdek and Borowska 2008). Dried berries are often used as food ingredients and in the preparation of fruit infusion or tea. Due to its pleasant fruit aroma, murta has a great potential for commercialization in dried state. Drying, as an appropriate postharvest technology, will prolong shelf life of the fruit and will preserve its quality and stability by lowering water activity through decrease of moisture content, thus avoiding spoilage and contamination during storage (Akpinar and Bicer 2005). Dehydrated murta berries can be considered an important source of vitamins, minerals and fibre and a probable component or ingredient of functional foods.

Knowledge of drying kinetics is required to design, optimise and control the drying process (Vega-Galvéz et al. 2008). To counter the undesirable effects of hot air-drying methods and to improve quality as well as nutritional value, vacuum drying is often attempted. It allows effective removal of moisture under low pressure (Jaya and Das 2003). Vacuum enhances the mass transfer because of an increased vapour pressure gradient between the inside and outside of the sample to dry and maintain a low temperature level essential for thermolabile products (Péré and Rodier 2002). Consequently, vacuum drying provides higher drying rate compared to conventional methods, lower drying temperature and offers an oxygen deficient processing environment (Wu et al. 2007). In addition, the analysis of the drying kinetics will help in designing suitable dryer with proper control of operating parameters, as well as in optimising the vacuum drying process with available basic data on drying kinetics. Vacuum drying has been performed successfully on other food materials, such as mango pulp (Jaya and Das 2003), coconut presscake (Jena and Das 2007), carrot and pumpkin (Cui et al. 2004; Arevalo-Pinedo and Murr 2006, 2007) and eggplants (Wu et al. 2007). However, prior to the drying study, a sorption analysis must be performed to know the moisture content of the product at equilibrium because drying behaviour is influenced by adsorption–desorption characteristics. This state of equilibrium results from multiple interactions on a microscopic scale, which is described by a relationship between the equilibrium water content of the product to be dried and the relative humidity of the atmosphere which surrounds it at a constant air temperature (Timmermann et al. 2001).

Simulation of the drying process through mathematical modelling is an important tool used to minimise operative problems such as product damage and excessive consumption of energy, among others (Babalis and Belessiotis 2004). The mathematical models used in literature fall into three categories, namely the theoretical, semi-theoretical and empirical. Theoretical models generally use the simultaneous heat and mass transfer equations with food properties such as particle geometry, shrinkage, moisture diffusion coefficient and critical moisture content. However, application of these models in the design or simulation of food dehydration may be more difficult and time consuming than empirical models that require little time and do not need assumptions of food properties. The empirical equations frequently used to model drying kinetics are Newton, Henderson–Pabis, Page, Modified Page, Wang–Singh, Logarithmic, Two-Terms, Modified Henderson–Pabis and other (Akpinar 2006). Most of these equations have been derived from Fick's second law of diffusion for different geometries (Doymaz 2007). Although empirical models would give good results for engineering applications in the food industry, they frequently do not allow the simulation of experiments carried out under conditions different to those used to identify the model parameters. In these cases, a theoretical model could be more appropriate. However, semi-theoretical models are most widely used, since they offer a compromise between theory and ease of application (Akpinar 2006).

The present study is focused on the modelling of hot air drying at 50, 60 and 70 °C of murta berries under vacuum and normal atmospheric pressure conditions. Diffusional and empirical models were developed to predict and compare the changes of moisture content occurring during the drying process under different pressure and temperature conditions.

Materials and Methods

Raw Material

Murta (*U. molinae* Turcz) or murtilla berries (Punucapa ecotype) were purchased in the city of Valdivia, location 38°48′30″S and 73°14′30″W, from merchants, who sell the wild berries plucked in the coastal forests in the surroundings of the city. Samples were selected to provide a homogeneous group, based on their colour, size and freshness according to visual analysis. They were stored at 4.5±0.2 °C (temperature) and 92.3±0.4% (relative humidity) in a refrigerator (Samsung SR-34RMB, Seoul, South Korea) for a maximum of 24 h before processing. The moisture content was determined according to Association of Official Agricultural Chemists (AOAC) methodology no. 934.06 (AOAC 1990), using a vacuum oven (JEIO Tech, OV-11, South Korea) at 70 °C for 72 h and an analytical balance (Mettler Toledo XS205 DU, Schwerzenbach, Switzerland) with an accuracy of ±0.01 g. Crude protein content was determined using the Kjeldahl method with a conversion factor of 6.25. Lipid content was analysed gravimetrically following Soxhlet extraction. Crude fibre was estimated by acid/alkaline hydrolysis of insoluble residue. Crude ash was estimated by incineration in a muffle furnace at 550 °C. Acidity was determined by the adapted AOAC methodology no. 942.15 (AOAC 1990), pH was measured using a potentiometer (Extech Instruments, Microcomputer pHVision 246072, Waltham, MA, USA), and soluble solids content was measured as saccharose using an Abbé refractometer (ATAGO, 1-T, Tokyo, Japan). All the analyses were made in triplicate and expressed in grams/100 g fresh product.

Determination of Equilibrium Moisture

Desorption isotherms were determined following the methodology recommended by the European Project COST Food Bioprocess Technol 90 (Spiess and Wolf 1983) at three working temperatures including 50, 60 and 70 °C. For this purpose, a known mass of sample, prepared in triplicate, was allowed to come into equilibrium with an atmosphere produced from a saturated salt solution having a known a_w including LiCl, $KC_2H_3O_2$, $MgCl_2$, K_2CO_3, $MgNO_3$, KI, NaCl, KCl and KNO_2 (all reagents were purchased from Merck KgaA, Darmstadt, Germany) (Kaymak-Ertekin and Gedik 2004). The sample and the salt solution were maintained separately within a sealed container, and the sample weight was taken every 15 days from the beginning of the test until it reached equilibrium (constant weight) (Vega-Gálvez et al. 2011). Then, moisture content reached in the desorption experiment was determined (AOAC 1990). The Guggenheim–Anderson–deBoer (GAB) model (Eq. 1), one of the most widely used models for description of equilibrium moisture isotherms of biological materials, was applied (Timmermann et al. 2001). In addition, this model is considered to have parameters based on physicochemical phenomena, such as monolayer moisture content (X_m) and the constants C and K, which are related to the first layer heat of sorption and multi-layer molecules with respect to the bulk liquid, respectively (Kaymak-Ertekin and Gedik 2004).

$$X_{we} = \frac{X_m\, C\, K\, a_w}{(1 - K\, a_w)[1 + (C - 1)K\, a_w]} \qquad (1)$$

where X_m is the monolayer water content, in grams water per gram dry matter (d.m.), and C and K are the GAB dimensionless parameters.

Drying Process

The drying experiment was carried out under steady-state condition (Lee and Kim 2009) at air temperatures of 50, 60, and 70 °C under vacuum (10 kPa absolute pressure) and atmospheric pressure using a laboratory oven (JEIO Tech, OV-11, South Korea) connected to a vacuum pump. The samples consisted of 30 individual berries arranged in a single thin layer on a chinaware drying tray and kept separated from each other within a grid (1.5 cm aperture) made of bamboo sticks of 3 mm diameter. Moisture losses of samples were recorded regularly for 24 h at half hour interval for the first 18 h and afterwards at 2-h interval until reaching a constant weight (equilibrium condition). An analytical balance (Mettler Toledo, XS205 DU, Schwerzenbach, Switzerland) with a precision of 0.01 mg was used for weight measurement. It was considered that the dry product has achieved equilibrium condition with the atmosphere inside the drying chamber at the end of each drying period.

Determination of Effective Moisture Diffusivity

Fick's second law of diffusion (Eq. 2) was used to interpret the drying process, since moisture diffusion is one of the main mass transport mechanisms that describes this process (Doymaz 2007). In this model, the dependent variable is moisture ratio (MR) which relates the gradient of sample moisture content in real time to both initial and equilibrium moisture content (Eq. 3).

$$\frac{\partial \mathrm{MR}}{\partial t} = D_{\mathrm{eff}} \frac{\partial^2 \mathrm{MR}}{\partial z^2} \qquad (2)$$

$$\mathrm{MR} = \frac{X_{wt} - X_{we}}{X_{wo} - X_{we}} \qquad (3)$$

where X_{wt} is the moisture content (in grams water per gram d.m.), X_{wo} the initial moisture content (in grams water per gram d.m.), X_{we} the equilibrium moisture content (in grams water per gram d.m.), D_{eff} the effective moisture diffusivity (in square meter per second), t the drying time (in seconds) and z the spatial dimension (in meter).

The mathematical solution of Fick's second law, when internal mass transfer is the controlling mechanism and one-dimensional transport in a sphere is assumed, is given by Eq. 4 (Crank 1975). For sufficiently long drying times, the first term in the series expansion gives a good estimate of the solution. In this case, a linear relationship between natural logarithm of MR and time is obtained, which can be used to determine effective moisture diffusivity (D_{eff}) according to Eq. 5.

$$\mathrm{MR} = \frac{6}{\pi^2} \sum_{j=0}^{\infty} \frac{1}{j^2} \exp\left[\frac{-j^2 D_{\mathrm{eff}} \pi^2 t}{a^2}\right] \qquad (4)$$

$$\mathrm{MR} = \frac{6}{\pi^2} \exp\left[\frac{-D_{\mathrm{eff}}\, \pi^2\, t}{a^2}\right] \qquad (5)$$

where a is the mean radius of the sphere (in meters) and j is the number of terms. The use of Eq. 5 is based on a constant D_{eff} assumption for each drying experiment and a linear behaviour between the mentioned variables. This hypothesis of isothermia is only a simple assumption, since drying is a complex process involving simultaneous heat and mass transfer, thus, all the complexity of the drying relies on the D_{eff} (Doymaz 2007).

The effective moisture diffusivity can be related to temperature by a simple Arrhenius-type relationship as given in Eq. 6. Activation energy was calculated by plotting the natural logarithm of D_{eff} against the reciprocal of absolute temperature.

$$D_{eff} = D_0 \exp\left(-\frac{E_a}{RT}\right) \tag{6}$$

D_o is the pre-exponential factor of the Arrhenius equation (in square meter per second), E_a the activation energy (in kilojoules per mole), R the universal gas constant (8.314 J/mol K) and T the absolute temperature (in Kelvin).

Mathematical Modelling of Drying Kinetics

Different mathematical models have been proposed to describe the drying kinetics of food and bioproducts, which derived from the diffusional model of Fick's second law for different geometries (Doymaz 2005; Akpinar 2006; Corzo et al. 2008). Table 1 presents the mathematical expressions of the selected models (Akpinar 2006) that were used to fit the experimental drying data of murta berries. The parameter k_i=1, 2…12 known as a kinetic parameter (1/min), could also be considered as a pseudo-diffusivity (Simal et al. 2005). The constants n_i=1, 2…12 and C are known as empirical parameters (dimensionless) that could depend on the existence of an external skin (Babalis and Belessiotis 2004). The shape parameter (α) is dimensionless and is related to the mass transfer rate at the beginning, e.g. the lower the α value, the faster the drying rate at the beginning (Corzo et al. 2008). The scale parameter β (in minutes) can be interpreted as a kinetic reaction constant and represents the time when concentration, in this case, $X_{wt}-X_{we}$ attains a value corresponding to 36.8% of $X_{wo}-X_{we}$ (Marabi et al. 2003). In addition, this study did not evaluate the influence of external skin, shrinkage and external resistance on kinetic and empirical parameters (Simal et al. 2005) since the mass transport inside the berry is assumed to be isotherm (thin thermal behaviour).

Statistical Analysis

For modelling the drying kinetics, the goodness of the fit between the predicted and experimental data was evaluated based on statistical analyses including sum squared error (Eq. 7), root mean squared error (Eq. 8) and chi-square (Eq. 9) (Akpinar 2006; Doymaz 2007). The effect of atmospheric and vacuum drying on water diffusion coefficients and empirical parameters was estimated using Statgraphics Plus® 5.1 (Statistical Graphics Corp., Herndon, VA, USA). The results were analysed by an analysis of variance (ANOVA). Differences between the media were analysed using the least significant difference test with a significance level of α=0.05 and a confidence interval of 95% (p<0.05). In addition, the multiple range test was used to demonstrate the existence of homogeneous groups.

$$\mathrm{SSE} = \frac{1}{N}\sum_{i=1}^{N}\left(\mathrm{MR}_{\mathrm{exp},i} - \mathrm{MR}_{\mathrm{calc},i}\right)^2 \tag{7}$$

$$\mathrm{RMSE} = \left[\frac{1}{N}\sum_{i=1}^{N}\left(\mathrm{MR}_{\mathrm{calc},i} - \mathrm{MR}_{\mathrm{exp},i}\right)^2\right]^{\frac{1}{2}} \tag{8}$$

Table 1 Mathematical models selected to describe the drying kinetics of murta berries

Model name	Model equation	References
Newton	$\mathrm{MR} = \exp(-k_1 t)$	Lemus-Mondaca et al. 2009
Henderson–Pabis	$\mathrm{MR} = n_1 \cdot \exp(-k_2 t)$	Akgun and Doymaz 2005
Page	$\mathrm{MR} = \exp(-k_3 t^{n_2})$	Gogus and Maskan 1999
Modified Page	$\mathrm{MR} = \exp(-(k_4 t)^{n_3})$	Toğrul and Pehlivan 2003
Wang–Singh	$\mathrm{MR} = k_5 t^2 + n_4 t + 1$	Uribe et al. 2011
Logarithmic	$\mathrm{MR} = C + n_5 \cdot \exp(-k_6 t)$	Akpinar 2006
Two terms	$\mathrm{MR} = n_6 \cdot \exp(-k_7 t) + n_7 \cdot \exp(-k_8 t)$	Lahsasni et al. 2004
Modified Henderson–Pabis	$\mathrm{MR} = n_8 \cdot \exp(-k_9 t) + n_9 \cdot \exp(-k_{10} t) + n_{10} \cdot \exp(-k_{11} t)$	Sacilik et al. 2006
Midilli–Kuçuk	$\mathrm{MR} = n_{11} \cdot \exp(-k_{12} t^{n_{12}}) + C\,t$	Vega-Gálvez et al. 2011
Weibull	$\mathrm{MR} = \exp\left(-\left(\frac{t}{\beta}\right)^{\alpha}\right)$	Corzo et al. 2008

$$\chi^2 = \frac{\sum_{i=1}^{N} \left(MR_{\exp,i} - MR_{\mathrm{calc},i}\right)^2}{N - m} \tag{9}$$

where MR_{exp} is the experimental moisture ratio, MR_{calc} the calculated moisture ratio, N the number of data values, m the number of constants and i the number of terms.

Results and Discussion

Samples Analyses and Equilibrium Moisture

Mean equatorial diameter of berries as measured by a Vernier caliper (Mitutoyo Digimatic Caliper, 500-144, People's Republic of China) was 11.48±0.67 mm, while the main axis diameter was 9.98±0.57 mm. Proximate analysis of murta berries, based on 100 g of fresh product, presented a moisture content of 77.40±1.01 g, crude protein (nitrogen x 6.25) of 1.00±0.30 g, total lipids of 0.80±0.10 g, crude fibre of 2.57±0.51 g, crude ash of 0.63±0.06 g and available carbohydrates (by difference) of 17.6±0.52 g. Soluble solid content was 16.48±0.19 °Brix, pH and tritimetric acidity were 3.82±0.27 and 1.007±0.141%, respectively. The results were comparable to those reported with murta *South Pearl-INIA* (Seguel and Montenegro 2008) and *Red Pearl-INIA* (Seguel and Montenegro 2010). It should be noted, however, that water content of the wild murta berries may vary during harvest season. The values of the desorption equilibrium moisture content, which is of particular interest for drying calculations, were obtained from a GAB model for the murta berries dried at 50, 60 and 70 °C (Fig. 1). The GAB constants used to calculate the corresponding equilibrium moisture content X_{we} at each drying temperature can be seen in Table 2.

Drying Kinetics

The mean total weight of murta berries did not exceed 21.0 g with a mean weight of single berry of 683.02±25.36 mg. Initial water content of the murta berries used in the drying experiments had a mean value of 79.89±1.08 g/100 g fresh product for experiments performed under vacuum and atmospheric pressure. The drying process was performed until equilibrium moisture was achieved. Beyond a drying period of 24 h, change in mass was merely in the order of hundredth milligram. Dry matter content for each individual berry could be calculated from sorption isotherm data as previously described, since the murta berries came to equilibrium water content at the end of experiments. Knowing initial weight of each berry, initial water content on a dry matter basis could also be determined.

In Fig. 1, the experimental drying curves, obtained from mean value of moisture ratio for experimental pressure and temperature conditions, are shown. The experimental results were consistent and showed as expected, that vacuum drying at any temperature was faster than drying under atmospheric pressure. Vacuum expands air and water vapour present in the food and creates a frothy or puffed structure, providing a large area-to-volume ratio for enhanced heat and mass transfer (Jaya and Das 2003). There is also a higher vapour pressure gradient between the inside and outside of the sample being dried (Péré and Rodier 2002). Consequently, with vacuum drying, it is possible to have a higher drying rate, a lower drying temperature and an oxygen-deficient processing environment (Wu et al. 2007). In Figs. 1 and 2, it can be observed that at higher air temperature, shorter drying time and higher drying rate are the results. The decrease in drying time and increase in drying rate with increase in the drying temperature have been observed by Akpinar (2006) for some vegetables and fruits; Babalis et al. (2006) for fig halves; Corzo et al. (2008) for mango slices; Vega-

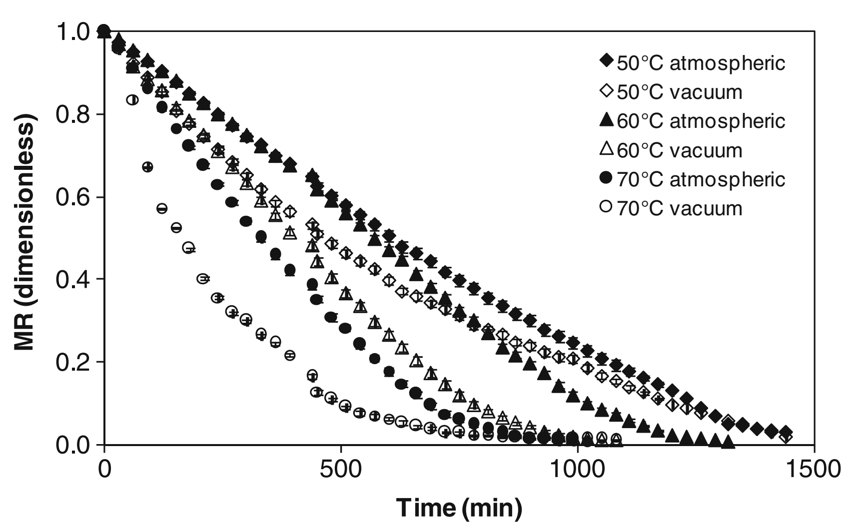

Fig. 1 Experimental drying curves at air temperatures of 50, 60 and 70 °C under vacuum and atmospheric pressure conditions

Table 2 GAB constants used to determine equilibrium moisture content for experiments under atmospheric pressure

Temperature	50 °C	60 °C	70 °C
X_m	0.077	0.077	0.078
C	5.680	6.540	6.998
K	0.965	0.978	0.922
r^2	0.9666	0.9637	0.9804
X_{we}	0.032	0.018	0.013
a_w	0.100	0.050	0.035

Gálvez et al. (2008) for red pepper and Lemus-Mondaca et al. (2009) for papayas.

As can be seen in Fig. 1, the drying curves for experiments at any particular temperature under either atmospheric pressure or vacuum conditions tend to meet at the end of the drying process. Since under vacuum conditions lower water content is reached earlier, drying rate is slower in that final phase. This can be explained by the lower driving force, apart from the fact that hardening of the outer layers of the berries would probably increase resistance to moisture diffusion inside the fruit. A slight change in drying rate is observed at around 450 min drying time, being more pronounced at 70 °C (Fig. 1), which may be due to changes in internal diffusion behaviour. It has been reported that particularly in hygroscopic products a third drying period may occur (Krischer and Kast 1978), as a result of release of bound water and levelling inside the product (Kröll 1978). There is also an increase of the solid–air interface temperature that reaches the hot air-drying temperature held constant and vapour pressure at solid surface tends to be equal to vapour pressure of drying air (Kröll 1978). This observation was not further investigated, since a more systematic approach would be needed. Moreover, considering that all the examined models showed low values of root mean square error (RSME), sum of square error (SSE) and chi-square, there was an indication that the observed change in drying rate was not significant.

The use of the experimental data or any of the developed models to calculate drying time to reach equilibrium moisture content gave results that differed slightly in magnitude. According to experimental data at 50 °C under atmospheric condition, drying time to reach equilibrium moisture was 1,435 min, while under vacuum a drying time of 1,409 min was observed. At 60 and 70 °C, the respective drying times for the respective pressure conditions were 1,237, 1,002, 930 and 900 min. This showed a clear effect of temperature but only a weak effect of vacuum that seemed to have more impact on the drying process of murta at 60 and 70 °C when MR is greater than 0.1; however, at 50 °C, MR should be over 0.2, as can be observed in Fig. 1. In a comparative study on different pretreatments of blueberries before performing convective drying at 70±0.2 °C and air velocity of 2.0±0.1 m/s, the untreated samples (control) needed 780 min to reach final water content (Vega-Gálvez et al. 2011). In the case of fig drying, Babalis et al. (2006) reported drying time of about 20 h to reach MR of 0.2 in a drying process at 65 °C and air velocity of 2.6 m/s.

Effect of Vacuum and Temperature on D_{eff}

Due to the fact that the relative humidity of the drying air at a higher temperature was less compared to that at a lower temperature, the difference in the partial vapour pressure between the berries and their surroundings was greater for the higher temperature drying environment. This resulted in a higher moisture transfer rate with the higher drying air temperature. Such an influence of drying air temperature on the drying rate was also noted in earlier researches (Lahsasni et al. 2004; Doymaz 2007).

The values of effective moisture diffusivity (D_{eff}) at different temperatures for drying under vacuum and atmospheric

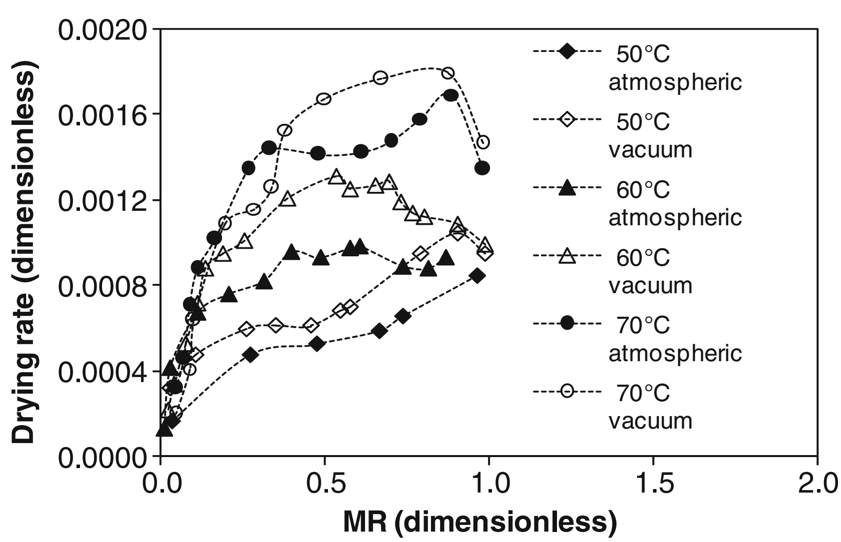

Fig. 2 Drying rate at air temperatures of 50, 60 and 70 °C under vacuum and atmospheric pressure conditions

pressure, obtained by using Eq. 5, are presented in Table 3. The average values of the effective diffusion coefficient of the murta berries in the drying process at 50–70 °C varied in the range of $3.10–11.30\times10^{-10}$ m^2/s. The values of D_{eff} increased progressively as the drying air temperature increased ($p<0.05$). It was also noted that effective diffusivity is always higher at any drying temperature under vacuum conditions. D_{eff} value doubled under vacuum conditions from 5.40×10^{-10} to 10.79×10^{-10} m^2/s when drying temperature increased from 50 to 60 °C ($p<0.05$). Under atmospheric conditions, similar increase occurred from 60 to 70 °C from 4.60×10^{-10} to 11.27×10^{-10} m^2/s. Several investigations carried out on fruits and vegetables under similar temperature conditions showed D_{eff} values to lie between 5.59 and 6.51×10^{-9} m^2/s for apricots (Toğrul and Pehlivan 2003); 1.33 and 3.36×10^{-9} m^2/s for figs (Babalis and Belessiotis 2004); 3.00 and 17.21×10^{-10} m^2/s for kiwi fruit (Simal et al. 2005); 4.27 and 13.00×10^{-10} m^2/s for okra (Doymaz 2005); 1.7 and 4.4×10^{-10} m^2/s for apples cv. Jonagold (Velić et al. 2004); 3.91 and 6.65×10^{-10} m^2/s for tomato (Doymaz 2007); 1.89 and 3.30×10^{-10} m^2/s for cherry laurel (Kaya and Aydin 2007); 0.48 and 2.02×10^{-10} m^2/s for apple cv. Red Delicious (Kaya et al. 2007); 7.02 and 37.82×10^{-9} m^2/s for red pepper var. Hungarian (Vega-Galvéz et al. 2008); 6.25 and 24.32×10^{-10} m^2/s for Chilean papaya (Lemus-Mondaca et al. 2009); and 6.92 and 14.59×10^{-9} m^2/s for radish slices (Lee and Kim 2009). Considering that the drying experiments for the murta berries were conducted under natural convection, it may be expected that moisture diffusion rate would increase if drying is performed under forced convection. The temperature dependence of D_{eff} can be described by an equation of the Arrhenius type as given in Eq. 6. Temperature has apparently a smaller effect on the moisture diffusivity in vacuum drying. This may be the result of a higher porosity of the vacuum-dried product provided by a large area-to-volume ratio (Jaya and Das 2003), increasing the vapour transfer rate associated to a higher vapour pressure gradient between the inside and outside of the sample being dried (Péré and Rodier 2002).

Table 3 Values of effective moisture diffusivity under vacuum and atmospheric pressure conditions during drying of murta at different temperatures

Temperature (°C)	Vacuum		Atmospheric	
	$D_{eff}\times10^{-10}$ (m^2/s)	r^2	$D_{eff}\times10^{-10}$ (m^2/s)	r^2
50	5.40±1.19a	0.93	3.10±0.56a	0.98
60	10.79±1.43b	0.93	4.60±0.88b	0.94
70	11.30±1.26b	0.99	11.27±0.88c	0.97

Different letters for the same column indicate that the D_{eff} values are significantly different ($p<0.05$)

The activation energy (E_a) and Arrhenius factor (D_o) were calculated from the inverse slope and intercept of the line plot of $\ln D_{eff}$ versus $1/T$. Activation energy can be interpreted as the energy barrier that must be overcome in order to activate moisture diffusion (Babalis and Belessiotis 2004). The E_a values of 34.30 ($r^2=0.8103$) and 59.27 kJ/mol ($r^2=0.94$) were obtained for drying process under vacuum and atmospheric pressure, respectively. In the case of vacuum drying, coefficient of determination was low, which may be due to the frothy or puffed structure with increased porosity reported by Jaya and Das (2003). The E_a values obtained in this study are similar to that of okra (51.26 kJ/mol) by Gogus and Maskan (1999) and green peppers (51.4 kJ/mol) by Kaymak-Ertekin (2002), but higher than that of lettuce and cauliflower leaves (19.82 kJ/mol) by Lopez et al. (2000), Uryani plums (24.83 kJ/mol) by Sacilik et al. (2006), apple slices (19.96–22.62 kJ/mol) by Kaya et al. (2007) and radish slices (16.49–20.26 kJ/mol) by Lee and Kim (2009).

The higher values of E_a for drying under atmospheric pressure and a high coefficient of determination r^2 indicate a clear effect of temperature on drying rate. In the case of vacuum drying of the murta berries, a linear relationship between natural logarithm of D_{eff} and reciprocal of temperature is not readily observed. This may be due to a higher increase rate of effective moisture diffusivity from 50 to 60 °C (99.8%) compared to that occurring between 60 and 70 °C (4.7%). When drying process takes place under vacuum conditions, a high diffusivity of moisture is achieved already at a temperature between 50 and 60 °C and only a slight increase occurred from 60 to 70 °C. Under atmospheric conditions, moisture diffusivity showed a rapid increase only at a higher temperature near to 70 °C. Since temperature may have a negative effect on some quality criteria of murta berries, drying of murta berries under vacuum conditions around 60 °C would be more appropriate than at 70 °C. This aspect should be duly considered in further studies on quality changes during drying of murta berries.

Kinetic Parameters of Drying Models

In this work, the models applied to fit the drying data of the murta berries were for most semi-theoretical. Table 4 shows the average values of the kinetic and empirical parameters $k_i=1, 2\ldots12$, $n_i=1, 2\ldots12$, C, α and β, obtained for all the proposed models. A tendency with increasing temperature was observed for each of these kinetic parameters, since in most models an increase in drying air temperature showed an increase in their values (Akpinar and Bicer 2005; Doymaz 2005; Akpinar 2006; Vega-Gálvez et al. 2008). These constants can therefore be considered to be directly proportional to the mass transfer coefficient. The Weibull model with its dimensionless shape parameter (α) and scale parameter (β) would give a good interpretation of the drying

Table 4 Kinetics and empirical parameters of the selected models used to simulate murta drying curves as related to each treatment

Empirical model		Vacuum			Atmospheric		
		50 °C	60 °C	70 °C	50 °C	60 °C	70 °C
Newton	k_1	0.0019±0.0002a	0.0037±0.0002b	0.0039±0.0002b	0.0013±0.0002a	0.0015±0.0002a	0.0040±0.0011b
Henderson–Pabis	n_1	1.3628±0.4287a	2.6051±0.7994a	1.5129±0.8586a	1.5745±0.5674a	2.6941±0.6432a	1.8785±0.5439a
	k_2	0.0022±0.0004a	0.0034±0.0002b	0.0047±0.0005c	0.0022±0.0005a	0.0037±0.0007b	0.0046±0.0006c
Page	n_2	1.1651±0.2875a	1.4651±0.3767a	1.0546±0.1133a	1.2832±0.2817a	1.5065±0.2799a	1.3856±0.2475a
	$k_3(\times10^{-1})$	0.0057±0.0004a	0.0013±0.0004b	0.0285±0.0018c	0.0021±0.0002a	0.0006±0.0001b	0.0027±0.0002c
Modified Page	n_3	1.1651±0.2875a	1.4651±0.3767a	1.0546±0.1133a	1.2832±0.2817a	1.5065±0.4799a	1.3856±0.2475a
	k_4	0.0016±0.0003a	0.0023±0.0002b	0.0039±0.0008c	0.0014±0.0002a	0.0017±0.0003a	0.0026±0.0003b
Wang–Singh	n_4	−0.0012±0.0004a	−0.0016±0.0003a	−0.0022±0.0004a	−0.0009±0.0002a	−0.0010±0.0004a	−0.0018±0.0004a
	$k_5(\times10^{-4})$	0.0035±0.0002a	0.0038±0.0004a	0.0041±0.0003a	0.0017±0.0002a	0.0019±0.0002a	0.0022±0.000a4
Logarithmic	C	−0.2036±0.0611a	−0.1659±0.0544a	0.0013±0.0004b	−0.9997±0.0437a	−0.7875±0.0411a	−0.0879±0.0054b
	n_5	1.2050±0.2600a	1.2480±0.2775a	1.0360±0.4459a	2.0140±0.3816a	1.8420±0.2772a	1.1680±0.3857a
	k_6	0.0011±0.0002a	0.0017±0.0002b	0.0044±0.0005c	0.0005±0.0002a	0.0007±0.0003a	0.0023±0.0004b
Two terms	n_6	−0.5791±0.1437a	0.6244±0.1088a	0.6052±0.1997a	0.5373±0.1115a	0.6216±0.1087a	0.5260±0.0943a
	k_7	0.0015±0.0002a	0.0018±0.0002a	0.0029±0.0003b	0.0017±0.0003a	0.0025±0.0002b	0.0044±0.0005c
	n_7	0.5225±0.1217a	0.5284±0.1784a	0.5137±0.1009a	0.5121±0.1604a	0.5162±0.1288a	0.5107±0.1427
	k_8	0.0015±0.0002a	0.0018±0.0004a	0.0029±0.0005b	0.0017±0.0002a	0.0025±0.0002b	0.0044±0.0003b
Modified	n_8	0.3549±0.06672a	0.4149±0.07172a	0.3461±0.0883a	0.3860±0.0447a	0.4173±0.04438a	0.4022±0.0511a
Henderson–Pabis	k_9	0.0017±0.0004a	0.0025±0.0004b	0.0044±0.0004c	0.0015±0.0001a	0.0018±0.0001b	0.0029±0.0002c
	n_9	0.3550±0.0894a	0.3795±0.1007a	0.3525±0.0816a	0.3683±0.1009a	0.3830±0.0968a	0.3755±0.1450a
	k_{10}	0.0017±0.0004a	0.0025±0.0004b	0.0044±0.0004c	0.0015±0.0001a	0.0018±0.0001b	0.0029±0.0002c
	n_{10}	0.3395±0.1108a	0.3433±0.0995a	0.3380±0.0933a	0.3473±0.1276a	0.3524±0.1097a	0.3413±0.1073a
	k_{11}	0.0017±0.0004a	0.0025±0.0004b	0.0044±0.0004c	0.0015±0.0001a	0.0018±0.0001b	0.0029±0.0002c
Midilli–Kuçuk	n_{11}	1.0101±0.0.2293a	0.9463±0.1028a	1.0235±0.1927a	0.9929±0.1765a	0.9563±0.1099a	0.9566±0.1230a
	n_{12}	0.9696±0.3482a	1.7439±0.4760a	1.043±0.4776a	1.1889±0.3762a	1.7676±0.3995a	1.5723±0.3218a
	$k_{12}(\times10^{-2})$	0.1600±0.03392a	0.0018±0.0004b	0.3400±0.05622c	0.0248±0.0043a	0.0008±0.0002b	0.0071±0.0015c
	$C\ (\times10^{-3})$	−0.0958±0.0068a	−0.0085±0.0016b	0.0045±0.0012c	−0.1588±0.0266a	−0.0451±0.0028b	−0.0056±0.0019c
Weibull	α	1.1651±0.2179a	1.4652±0.2002b	1.0546±0.1890c	1.2832±0.2896a	1.5065±0.2999b	1.3856±0.1977c
	β	609.18±18.56a	439.21±21.35b	259.30±16.66c	727.68±29.01a	605.69±26.39b	381.23±11.26c

Different letters in the same line indicate that the values are significantly different ($p<0.05$)

kinetic of the murta berries. As can be seen in Table 4, the shape parameter ranged from 1.05 to 1.51 and indicated higher drying rate for vacuum drying than for atmospheric drying. The scale parameter (β) ranged from 259.30 to 727.68 min showing relatively long drying period compared to drying under forced convection, which cannot be implemented for vacuum drying. Values of α in a range from 1.22 to 1.41 and values of β from 98.88 to 244.37 min were found in the case of pepino fruit dried at a constant air flow rate of 2.0±0.1 m/s at temperatures between 50 and 90 °C. This shows that for a similar initial drying rate, forced convection would most probably reduce drying time. A $p>0.05$ was obtained from the ANOVA on the averages of parameters $n_i=1, 2\ldots12$, α and C, suggesting that there was no significant influence of the treatments (drying in atmospheric pressure and under vacuum) used on these empirical parameters (Table 4). The same statistical evaluation (ANOVA) was carried out on the averages of the kinetic parameters $k_i=1, 2\ldots12$ and β (Table 4), obtaining a $p<0.05$, which suggests a significant influence of the treatments used on these kinetics parameters.

Figure 3 compares the experimental and the predicted moisture ratios of the Midilli–Ku uk and Weibull models as a function of drying time in processing of murta berries at 50,

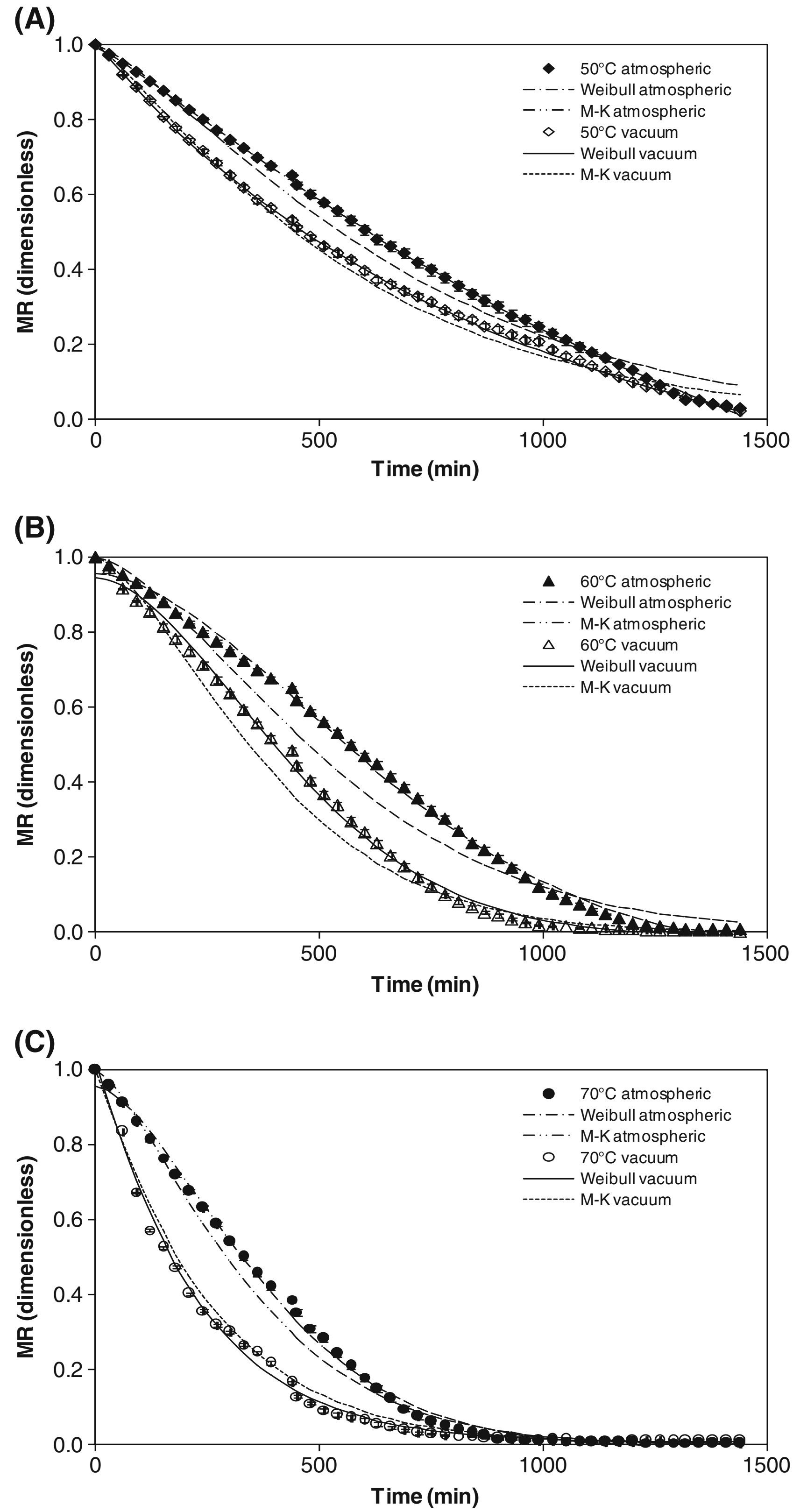

Fig. 3 Experimental and calculated drying curves for murta under atmospheric and vacuum conditions at (**a**) 50, (**b**) 60 and (**c**) 70 °C for Midilli–Ku uk and Weibull models

60 and 70 °C under vacuum and atmospheric pressure conditions, respectively. As can be seen, the proposed models provided conformity between experimental and predicted moisture ratios. The Page and modified Page models are similar to the Midilli–Ku uk model. However, the Weibull model shows at 60 °C for both vacuum and atmospheric

conditions a slight deviation to experimental results at the beginning of drying process. The modified Page model, which is an alternate form of the Weibull model, has been applied with good results by many authors in describing drying kinetics of different foods (Toğrul and Pehlivan 2003; Lahsasni et al. 2004; Akpinar and Bicer 2005; Doymaz 2005; Simal et al. 2005; Akpinar 2006; Vega-Gálvez et al. 2008). Considering that an important aspect of the drying kinetic models is to predict drying rate and drying time, the Weibull can be used with good accuracy in the case of murta berries.

Statistical Analysis on the Drying Models

Table 5 shows the summary of the statistical evaluation for the theoretical drying kinetic models for each test applied. Based on this evaluation, a good fit was observed for most of the models. Low SSE (≤0.009), RMSE (≤0.090) and χ^2 (≤0.009) values are considered decisive for a good model. In addition, the criterion of r^2≥0.90 is also considered to evaluate if models show a good fit quality to the experimental data. From the selected semi-theoretical models, the Weibull model had statistically the best quality of fit for all experimental drying conditions. Midilli–Ku uk, Page and modified Page models are also statistically solid models (Table 5). Then, these models could be a good option to fit the experimental data. The highest values for SSE (≤0.5915), RSME (≤0.7691) and χ^2 (≤0.6166) were computed in the case of the empirical Wang–Singh model, for drying at 70 °C under vacuum. This empirical model seemed less appropriate to simulate the entire drying process of murta berries. The logarithmic and the Henderson–Pabis models, and in some cases the Newton and the two terms models, also have SSE, RSME and χ^2 values which make them weaker models to represent the drying kinetics of murta berries.

Table 5 Statistical tests for each empirical model as related to each treatment

Model name	Statistics	Vacuum			Atmospheric		
		50 °C	60 °C	70 °C	50 °C	60 °C	70 °C
Newton	SSE	0.0037	0.0260	0.0007	0.0045	0.0094	0.0152
	RMSE	0.0610	0.1612	0.0261	0.0672	0.0967	0.1233
	χ^2	0.0039	0.0271	0.0007	0.0047	0.0098	0.0159
Henderson–Pabis	SSE	0.0102	0.0429	0.0023	0.0288	0.0698	0.0232
	RMSE	0.1011	0.2072	0.0475	0.1697	0.2643	0.1523
	χ^2	0.0107	0.0448	0.0023	0.0300	0.0728	0.0242
Page	SSE	0.0005	0.0019	0.0006	0.0014	0.0031	0.0010
	RMSE	0.0225	0.0431	0.0242	0.0369	0.0552	0.0312
	χ^2	0.0005	0.0019	0.0006	0.0014	0.0032	0.0010
Modified Page	SSE	0.0005	0.0019	0.0006	0.0014	0.0031	0.0010
	RMSE	0.0225	0.0431	0.0242	0.0369	0.0552	0.0312
	χ^2	0.0005	0.0019	0.0006	0.0014	0.0032	0.0010
Wang–Singh	SSE	0.0013	0.0596	0.5915	0.0004	0.0019	0.3484
	RMSE	0.0366	0.2441	0.7691	0.0189	0.0435	0.5902
	χ^2	0.0014	0.0621	0.6166	0.0004	0.0020	0.3632
Logarithmic	SSE	0.0737	0.0356	0.0066	0.1967	0.1671	0.0589
	RMSE	0.2715	0.1886	0.0811	0.4435	0.4087	0.2428
	χ^2	0.0768	0.0371	0.0069	0.2051	0.1742	0.0614
Two terms	SSE	0.0019	0.0100	0.0079	0.0107	0.0297	0.0201
	RMSE	0.0436	0.1001	0.0888	0.1033	0.1725	0.1419
	χ^2	0.0020	0.0104	0.0082	0.0111	0.0310	0.0210
Modified H-P	SSE	0.0014	0.0063	0.0004	0.0056	0.0113	0.0044
	RMSE	0.0368	0.0793	0.0198	0.0748	0.1063	0.0664
	χ^2	0.0014	0.0066	0.0004	0.0058	0.0118	0.0046
Midilli–Kuçuk	SSE	0.0005	0.0019	0.0006	0.0014	0.0031	0.0010
	RMSE	0.0225	0.0431	0.0242	0.0369	0.0552	0.0312
	χ^2	0.0005	0.0019	0.0006	0.0014	0.0032	0.0010
Weibull	SSE	0.0001	0.0009	0.0003	0.0000	0.0009	0.0006
	RMSE	0.0100	0.0301	0.0173	0.0067	0.0292	0.0248
	χ^2	0.0001	0.0009	0.0003	0.0000	0.0009	0.0006

RMSE root mean square error, *SSE* sum of square error

Conclusions

This study showed that modelling of vacuum and atmospheric drying of murta berries under natural convection allows a comparison of the basic dehydration characteristics of the berry. According to statistical tests applied, the Weibull and Midilli–Ku uk models showed the best fit among all selected models, having the lowest values for SSE, RMSE and χ^2. Page and modified Page models are also suitable to describe the drying kinetics of the murta berries. Drying time decreased with an increase in drying temperature from 1,500 to 1,000 min and is shorter under vacuum conditions from 1,500 to 800 min. The effect of vacuum is more relevant at moisture ratio over 0.2. Effective moisture diffusivity, as quantified experimentally, also showed that drying of murta could be performed under vacuum condition at 60 °C (D_{eff}=10.8×10^{-10} m^2/s), since comparable diffusivity value to atmospheric drying at 70 °C (D_{eff}= 11.27×10^{-10} m^2/s) is obtained.

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