Studii şi Cercetări Ştiinţifice
Chimie şi Inginerie Chimică, Biotehnologii, Industrie Alimentară

2018, **19** (3), pp. 243 – 255

Scientific Study & Research
Chemistry & Chemical Engineering, Biotechnology, Food Industry

ISSN 1582-540X

ORIGINAL RESEARCH PAPER

CONVECTIVE AIR DRYING CHARACTERISTICS OF GROUND MACADAMIA NUTS

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Received: December, 31, 2017
Accepted: July, 06, 2018

Abstract: Convective hot air drying of ground macadamia (particle size corresponding to 1.29 ± 0.11 mm) was investigated in the temperature and air velocity ranges of 40 to 70 °C and 0.7 to 2.5 $m \cdot s^{-1}$, respectively. Page and two-term exponential models were identified in this study as the best semi-theoretical models to describe the thin layer drying of ground macadamia nuts. Both experiments and models revealed that shorter drying times correspond to higher temperatures and smaller granulometries. The effective diffusivity of moisture transfer varied from 3.7 x 10^{-9} to 4.4 x 10^{-9} $m^2 \cdot s^{-1}$ and activation energy from 9.57 $kJ \cdot mol^{-1}$ to 18.57 $kJ \cdot mol^{-1}$ over the temperature and air velocity ranges used in this study. It was also found that when macadamia nuts are reduced to particulate form, diffusivity and activation energy increase.

Keywords: *convective air drying, dehydration, drying rate, macadamia, modeling, nuts*

INTRODUCTION

Macadamia (*Macadamia integrifolia*) tree, originated from the Australian tropical forests belongs to the family of Proteaceae [1]. Australia, Guatemala, Brazil Costa Rica and South Africa are the world largest exporters of macadamia nuts. The South African macadamia industry has grown into a major world force, competing with Australia in terms of being the largest producer. It is arguably the fastest growing tree crop industry in South Africa from 1211 tons of nut in shell in 1991 to 38000 tons in 2016 [2].

Every part of the macadamia crop can be used. The outer husk is combined with other organic matter to produce an excellent manure for young trees. The hard shell sustains high temperatures when burning and is mostly used as a fuel for firing furnaces. When the shell is ground to a fine powder the resulting granules are extremely hard. This powder can be used as an industrial abrasive and is superior to sand for sand blasting. It is even marketed by the cosmetics industry as the active ingredient in facial skin scrub [3]. The kernels of the macadamia nut are mainly consumed as snack food. Macadamia may also be used in baked goods and confectionery [3]. The nuts are rich in mono-unsaturated oils which have highly valued uses in the cosmetic and pharmaceutical industries.

Agricultural products, including macadamia nuts, are subjected to drying. This unit operation, defined as the removal of relatively small amounts of a liquid from a process stream, serves the purpose of preserving and extending the shelf-life of agricultural products while reducing packaging, storage and transportation costs [4]. In many regions around the world, crops are still dried by direct exposure to the sun. This technique, although simple and economic, suffers from some disadvantages including the risk of product contamination from dust and insects as well as food deterioration resulting from enzyme and microbial activity [5]. To overcome these shortcomings, many types of dryers have been proposed. One such is the convective air dryer. The optimal design of industrial dryers requires a better understanding of the dehydration process as obtained from experimental data as well as relevant modeling studies. The same apply to the design and optimization of processes involving at least one drying step [4]. Furthermore, there is need for experimental data to test or validate existing as well as newly developed models related to crop drying. Bearing this in mind, researchers reported in the literature the drying behavior of various fruits and vegetables used as food or mere industrial crops. Examples include the hot air drying characteristics of garlic [6], red pepper [7, 8], okra [9, 10], dill as well as parsley leaves [11], tomato [12], cornelian cherry fruits [13], water chestnut [14], sweet cherry [15], rapeseed [16], passion fruit peel [17], sorbus fruits [18], taro [19], elephant foot yam [20] and grape [21]. With regard to macadamia nuts drying, Palipane and Driscoll [22] investigated the effect of temperature and relative air humidity on the convective air drying behavior of in-shell nuts on one hand and kernels on the other. They successfully fitted their experimental data to the two-term exponential drying model. The major finding of this work was to have established that at lower moistures, kernels' drying was faster when in-shell nuts were investigated than when extracted kernels were used. Using a different technique, *i.e.* microwave assisted convective air drying, Silva *et al.* [1] examined the effect of temperature on the drying kinetics. This technique led to shorter processing times and better macadamia kernel nutritional quality than those obtained after convective air drying. Their experimental data were accurately correlated by means of Fick's second law. Poogungploy *et al.* [23] recommended microwave assisted hot air drying by surface temperature control at 60 °C with hot air at this same temperature. An investigation by Chung and Furutani [24] revealed that microwave power level was the most important factor affecting the quality of the processed kernels. Recent studies showed that radiofrequency (RF) process could provide rapid, uniform, and quality-acceptable drying technology for macadamia nuts [25 – 28]. Cykler [29] and Borompichaichartkul [30] reported promising results when combining heat pump and hot drying. They observed shorter drying times than in conventional hot air drying process and natural quality preservation of macadamia nuts. Jongjaipak *et al.* [31] combined electric field and convective hot air drying of macadamia what lead to higher drying rate, lower specific energy consumption and lower peroxide value. However, the electric field negatively affected nut colour.

A search of the literature revealed no previous study on ground macadamia nuts, which are relevant to some industrial processes such as oil extraction. In effect, temperature, moisture content and particle size particle are very important parameters for oil extraction from grains. This absence of data motivated the present study, aimed at investigating the effect of selected operating parameters (*i.e.*, temperature and air velocity) on hot air drying kinetics of ground macadamia. In the present study, moisture ratios during ground macadamia nuts drying are reported at 3 different temperatures and three air velocities. Diffusivities and activation energies calculated from experimental data are compared with similar data found in the literature.

MATERIALS AND METHODS

Materials

Macadamia nuts used in this study were harvested from a domestic tree in Tongaat, a small town on the North Coast of the South African KwaZulu-Natal province. After open air sun-drying, the nuts were manually dehusked, unshelled and ground using a laboratory mortar and pestle. Particle size analyses were undertaken by means of a shaker. For experimentation, the particle size range corresponding to 1.29 ± 0.11 mm was considered. Ground macadamia samples were stored in sealed aluminum bags under refrigeration at approximately 4 °C until needed. The initial moisture content of ground macadamia samples, determined by the oven method [12] at 105 °C for 24 h, was found to be 53 ± 0.5 % on a dry basis.

Experimental set up

The drying behavior of ground macadamia nuts was investigated in a laboratory convective air dryer supplied by GUNT Geratëbau (model CE 130, manufactured in Germany). The experimental set up, schematically represented in Figure 1, consisted of a centrifugal fan for air flow supply, an air filter, an electric heater.

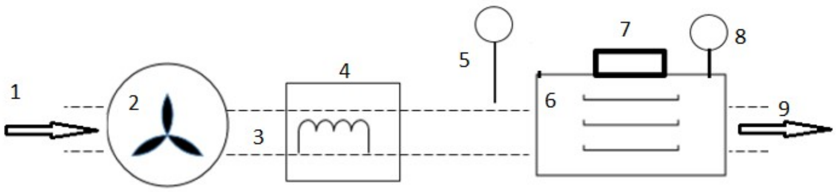

Figure 1. *Schematic diagram of the convective air dryer 1: fresh air inlet, 2: fun, 3: flow channel, 4: heating element and control cabinet, 5: anemometer, temperature probe and humidity sensor, 6: drying chamber with three material holders (trays), 7: digital scale, 8: anemometer, temperature probe and humidity sensor, 9: exit air*

The direction of airflow was horizontal over the drying tray. One interesting feature of the dryer is the presence of a glass viewing window, allowing a visual observation of the samples during dehydration experiments. Controllers were fitted on the equipment for both air flow and temperature. Temperatures and velocities were respectively measured by a Pt 100 and an anemometer incorporated to the equipment. Uncertainties in measuring temperatures and velocities were estimated as ± 1 °C and ± 0.1 $m \cdot s^{-1}$. The change of mass with time was monitored by a Kern^{TM} balance (model KB; manufactured by Kern & Sohn in Germany; accuracy: ± 0.00005 g). All experimental parameters, *i.e.* time, temperature, mass, and air relative humidity were logged on a computer through customized in-house Microsoft Windows-based software provided by GUNT Geratëbau (Germany) as part of the equipment.

Experimental procedure

Woven wire mesh sieves of various aperture sizes were used to determine the particle size distribution of ground macadamia nuts. These sieves were mounted on a shaker which assisted in product size segregation. Sieve analysis was selected as the method of particle size distribution since this method is applicable for granular materials in the particle size range between 32 and 5600 µm [32].
Convective air drying experiments were undertaken to study the temperature and air velocity on the dehydration behavior of ground macadamia nuts. Experimental runs were performed in triplicate at three different temperatures (40, 50, and 60 °C), three different air velocities (0.7, 1.5, and 2.3 $m \cdot s^{-1}$). The particle size of samples used in this study was 1.29 ± 0.11 mm. The fan was first switched on and the air velocity set at the desired value. The temperature was subsequently set at the value of interest as well. After setting the air velocity and temperature at the desired values, the fan and the heater were switched on and the dryer was allowed to run for one hour before spreading a single layer of ground macadamia nuts on the drying plate. The balance was tarred and the change of mass was recorded by the software and displayed in real time on the computer monitor, along with time and other experimental parameters. Once the weight became constant over time (within 5 %), the drying process was considered complete. Results from repeated experiments (three times for each set of variables) did not exhibit notable discrepancies.

Modeling

Moisture content, moisture ratio and drying rate calculation

Drying data are generally presented in terms of moisture content, moisture ratio as well as drying rate as dependent variables and time being the independent variable.

Moisture content (M_t) at any time, expressed as kg H_2O / kg dry mass, can be calculated as a function of time (t in minutes) as follows:

$$M_t = \frac{m_w}{m_{db}} = \frac{m_t - m_{db}}{m_{db}} \tag{1}$$

where: m_{db} is the mass of the dried sample, and m_t is the mass of the sample inside the drying chamber at time t.

Moisture ratio (MR) is determined as a dimensionless value using the following equation:

$$MR = \frac{M_t - M_e}{M_o - M_e} \tag{2}$$

Assuming that the equilibrium moisture (M_e) content is by far less than both the moisture content at time t (M_t) and the initial moisture content (M_o), the following MR expression can be obtained:

$$MR = \frac{M_t}{M_o} \tag{3}$$

The drying rate (DR) can be calculated using the following equation:

$$DR = \frac{M_t - M_{t+dt}}{dt} \tag{4}$$

where: M_{t+dt} represent moisture content at time $t + dt$, whereas dt is the time interval between t and $t + dt$.

It is generally accepted that diffusion is the main mechanism governing the transport of moisture to the surface to be evaporated during drying. Hence the widespread use of Fick's equation in the literature to gain insights into the drying process of various agricultural products, assuming a spherical shape [11, 33]:

$$MR = \frac{6}{\pi^2}\sum_{n=1}^{\infty}\frac{1}{n^2}\exp\left(-\frac{\pi^2 n^2 D_{eff} t}{r^2}\right) \tag{5}$$

where: D_{eff} is the effective diffusivity in $m^2 \cdot s^{-1}$, r is the radius, t is the time.

Taking into account the first term of the above general series solution of Fick's second law, the following expression can be obtained for MR:

$$MR = \frac{6}{\pi^2}\exp\left(\frac{\pi^2 D_{eff} t}{r^2}\right) \tag{6}$$

Many other empirical and semi-empirical models have been proposed in the literature to describe the change of moisture ratio as a function of time. These thin-layer drying models presented in Table 1 are usually fitted to experimental drying data.

Table 1. *Thin-layer drying models for drying data correlation 1 (adapted from a publication by Mujaffar and Loy [34])*

No	Model	Equation
1.	Newton	$MR = exp(-kt)$
2.	Page	$MR = exp(-kt^n)$
3.	Modified Page	$MR = exp(-kt)^n$
4.	Henderson and Pabis	$MR = a\ exp(-kt)$
5.	Modified Henderson and Pabis	$MR = a\ exp(-kt) + b\ exp(-gt) + c\ exp(-ht)$
6.	Logarithmic	$MR = a\ exp(-kt) + c$
7.	Two-term	$MR = a\ exp(-k_1 t) + b\ exp(-k_2 t)$
8.	Two-term exponential	$MR = a\ exp(-k t) + (1-a)\ exp(-kat)$
9.	Wang & Singh	$MR = 1+at+bt^2$
10.	Verma	$MR = a\ exp(-kt)+(1-a)\ exp(-gt)$
11.	Hii	$MR = a\ exp(-kt^n) + c\ exp(-gtn)$
12.	Midilli	$MR = a\ exp(-kt^n) + b\ t$
13.	Peleg	$MR = 1 - (x/(a+bx))$
14.	Weibull distribution	$MR = a - b\ exp(-kt^n)$
15.	Diffusion approach	$MR = a\ exp(-kt)+(1-a)\ exp(-kbt)$
16.	Aghbashlo *et al.*	$MR = -k_1 t / (1 + k_2 t)$
17.	Logistic	$MR = a_0 / ((1 + a\ exp(kt))$
18.	Jena and Das	$MR = a\ exp(-kt + bt^{1/2}) + c$
19.	Demir *et al.*	$MR = a\ exp(-(kt)^n) + c$
20.	Simplified Fick's diffusion equation	$MR = a\ exp(-c\ (t/L^2))$
21.	Modified Page equation-II	$MR = exp(-k\ (t/L^2))^n$
22.	Alibas	$MR = a\ exp(-kt^n + b\ t) + g$
23.	Ademiluyi *et al.*	$MR = a \cdot exp(-(kt)^n)$

Diffusivity and activation energy

From isothermal drying data, effective diffusivity (D_{eff}) can be obtained from equations 7 and 8. The plot of natural logarithm of MR versus time should yield a straight line displaying a slope (k) given by the following equation:

$$k = \frac{\pi^2 D_{eff}}{r^2} \tag{7}$$

which can also be written as:

$$D_{eff} = \frac{kr^2}{\pi^2} \tag{8}$$

It is also common practice to determine the activation energy associated with the drying process from temperature-dependence of effective diffusivity (D_{eff}) through the following Arrhenius-like equation:

$$D_{eff} = D_o\ \exp\left(-\frac{E_a}{RT}\right) \tag{9}$$

where D_o stands for the diffusivity value for an infinite moisture content in $m^2 \cdot s^{-1}$ and E_a is the activation energy in $J \cdot mol^{-1}$. R and T are the universal gas constant in $J \cdot mol^{-1} \cdot K^{-1}$ and the temperature in K, respectively. In order to obtain the activation energy value, a plot of ln (D_{eff}) should be plotted against ($1/T$). The slope and the intercept of this straight line allow calculating D_o and E_a.

Statistical analysis

Once models are fitted to experimental data, statistical parameters can be used to assess their performance, *i.e.* the goodness of fit. For this purpose, most researchers reporting drying studies for agricultural crops used the coefficient of determination (R^2), reduced sum square error (SSE), root mean square error ($RMSE$) and the reduced chi-square (χ^2) given by the following equations:

$$R^2 = \left[\frac{n\sum_{i=1}^{n} MR_{\exp,i} . MR_{cal,i} - \sum_{i=1}^{n} MR_{\exp,i} \cdot \sum_{i=1}^{n} MR_{cal,i}}{\sqrt{n\left(\sum_{i=1}^{n} MR_{\exp,i}^2\right) - \left(\sum_{i=1}^{n} MR_{\exp,i}\right)^2} \sqrt{n\left(\sum_{i=1}^{n} MR_{cal,i}^2\right) - \left(\sum_{i=1}^{n} MR_{cal,i}\right)^2}} \right]^2 \quad (10)$$

$$SSE = \frac{\sum_{i=1}^{n} \left(MR_{\exp,i} - MR_{cal,i}\right)^2}{n} \quad (11)$$

$$RMSE = \sqrt{\frac{\sum_{i=1}^{n} \left(MR_{\exp,i} - MR_{cal,i}\right)^2}{n}} \quad (12)$$

$$\chi^2 = \frac{\sum_{i} \left(MR_{\exp,i} - MR_{cal,i}\right)^2}{n - c} \quad (13)$$

where $MR_{exp,i}$ and $MR_{cal,i}$ denote the experimental and the calculated moisture ratio, respectively, while n is the number of experimental data points, and c represents the number of constants in the equation representing the model.

A model is considered better than another if it has a higher coefficient of determination (R^2) and lower values of reduced sum square error (SSE), root mean square error ($RMSE$) and the reduced chi-square (χ^2). In this study, model constants were determined through Differential Evolutionary optimization strategy implemented in Matlab by minimizing the sum square error given by equation (10).

RESULTS AND DISCUSSION

Effect of process parameters

Convective hot air drying experiments of ground macadamia (particle size: 1.29 ± 0.11 mm) were undertaken at 40, 50 and 60 °C and 0.7, 1.5 and 2.5 $m \cdot s^{-1}$. Furthermore, experimental data obtained in this study were fitted to two different models, *i.e.* Page and two-term exponential models. Both experimental and modeling results (for the two-term exponential model only) in terms of moisture ratio versus time are presented in Figure 2. These drying curves show that higher temperatures as well as higher air velocities lead to shorter drying times. It is interesting to note that the effect of air velocity on drying time is very weak as visible in Figure 2. The same trends were observed by other researchers who reported experimental data on convective air drying of food and bio-products [7, 8]. Drying times obtained in this study are lower than those reported by Palipane and Driscoll [22] who investigated whole macadamia nuts. This can be attributed to the increased specific surface area resulting in enhanced surface moisture removal.

To gain some insights into the dehydration mechanism, the drying rate curves of which a sample is shown in Figure 3 were plotted and examined. The trend exhibited by these curves suggests that dehydration mostly occurs in the falling rate period. The straight line portion of the drying rate curves covers more than 50 % of the drying time. For this reason, it can be stated that the drying process of ground macadamia nuts is mostly governed by internal mass transfer.

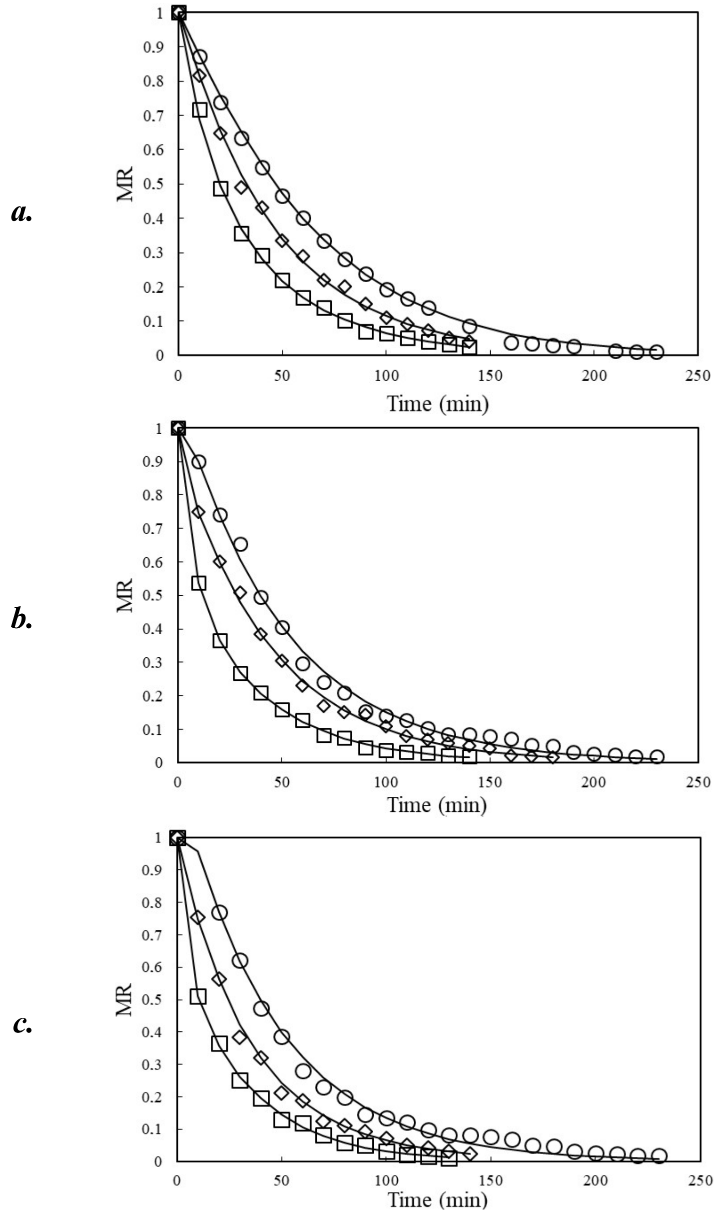

Figure 2. *Variation of ground macadamia moisture ratio (MR) with time (t) at 40 °C (o), 50 °C (◊) as well as 60 °C (□) and different velocities: a) 0.7 m·s^{-1}; b) 1.5 m·s^{-1}; c) 2.3 m·s^{-1} (values predicted by the two-term exponential model)*

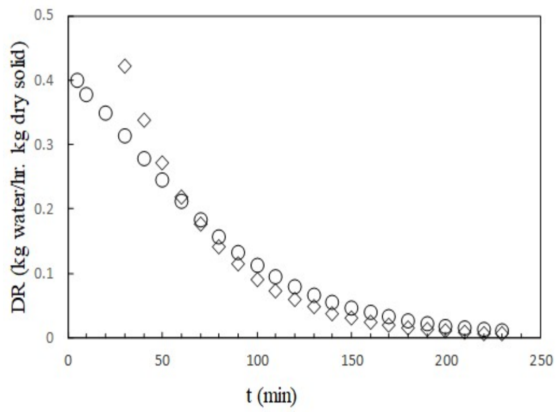

Figure 3. *Drying rate (DR) curves at 40 °C and 0.7 m·s^{-1} (o), 2.3 m·s^{-1} (◊) air velocities*

Evaluation of the models

As part of this study, attempts were made to correlate experimental drying data using all the models presented in Table 1. Models were evaluated on the basis of the statistical parameters given by equations 10 to 13. Calculated values of these parameters, along with model constants are provided in Table 2 for Page model and in Table 3 for two-term exponential model which emerged as the best semi-theoretical models to describe the drying of ground macadamia nuts.

Table 2. *Constants for Page model derived in this study for different temperatures and air velocities**

T [°C]	v [m·s^{-1}]	k [min^{-1}]	n	R^2	SSE	RMSE	χ^2
40	0.7	0.0105	1.0950	0.9991	0.00012	0.01076	0.00013
	1.5	0.0138	1.0649	0.9934	0.00059	0.02424	0.00064
	2.5	0.0181	1.0124	0.9908	0.00068	0.02606	0.00074
50	0.7	0.0191	1.0284	0.9978	0.00019	0.01375	0.00022
	1.5	0.0325	0.9191	0.9981	0.00014	0.01197	0.00016
	2.5	0.0339	0.9519	0.9974	0.00022	0.01475	0.00025
60	0.7	0.0509	0.8663	0.9984	0.00013	0.01127	0.00015
	1.5	0.1254	0.6936	0.9994	0.00004	0.00659	0.00005
	2.5	0.1172	0.7261	0.9985	0.00011	0.01043	0.00013

Table 3. *Parameters for the two-term exponential model (a, b, k_1 and k_2) derived in this study for different temperatures and air velocities**

T [°C]	v [m·s^{-1}]	a	b	k_1 [min^{-1}]	k_2 [min^{-1}]	R^2	SSE	RMSE	χ^2
40	0.7	8.1091	-7.1091	0.0228	0.0242	0.9992	0.00008	0.00915	0.00010
	1.5	-0.0975	1.0975	2.9795	0.0198	0.9958	0.00036	0.01900	0.00043
	2.5	1.1900	-0.1900	0.0218	7.3979	0.9958	0.00037	0.01924	0.00045
50	0.7	-0.0140	1.0140	4.7761	0.0218	0.9980	0.00017	0.01291	0.00023
	1.5	0.9352	0.0648	0.0223	4.1763	0.9983	0.00013	0.01137	0.00016
	2.5	0.1904	0.8096	0.0181	0.0316	0.9977	0.00020	0.01423	0.00028
60	0.7	0.6862	0.3138	0.0236	0.0773	0.9989	0.00010	0.00987	0.00013
	1.5	0.5797	0.4203	0.0258	0.1535	0.9997	0.00002	0.00484	0.00003
	2.5	0.6449	0.3551	0.0299	0.2426	0.9993	0.00005	0.00699	0.00007

Of these models, the two-term exponential model was the most accurate approach for drying data correlation. This finding is consistent with the study related to whole macadamia nuts as reported by Palipane and Driscoll [22]. With regard to the best performing model for moisture ratio, *i.e.* the two-term exponential model, the coefficient of determination (R^2), reduced sum square error (*SSE*), root mean square error (*RMSE*) and the reduced chi-square (χ^2) were found in the ranges (0.996 - 0.999), (0.00002 - 0.0004), (0.005 - 0.02), and (0.00003 - 0.00045) respectively.
The good quality of fit can also be visually confirmed by examining Figure 2 on one hand and Figure 4 on the other hand.

a.

b.

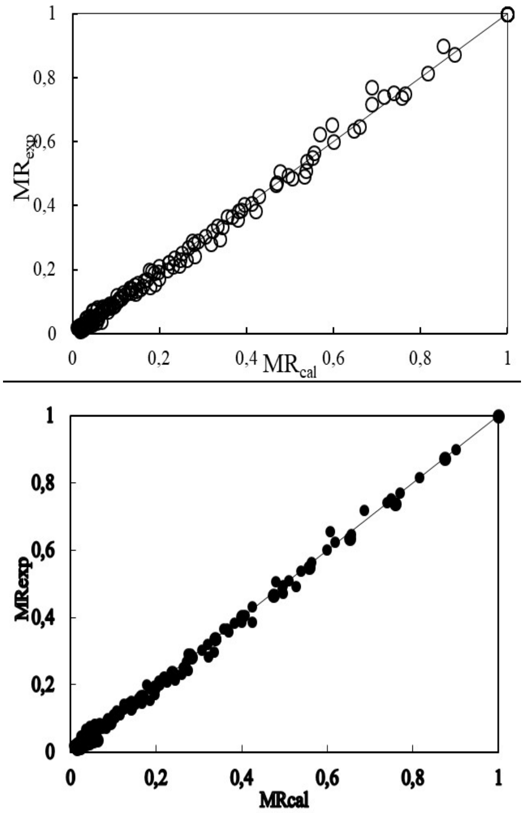

Figure 4. *Comparison between all experimental moisture ratios (MR_{exp}) and those calculated (MR_{cal}) by Page model (a) and by two-term exponential model (b)*

Most points appearing in Figure 4 remain close to the diagonal line. It is an indication of good agreement between experimental data and those calculated using the two models.

Moisture diffusivity and activation energy

Given that internal resistance to mass transfer governs the drying of ground macadamia, Fick's second law was used to calculate the effective diffusivity. Following the procedure outlined in section 2 of this article, effective diffusivity values were estimated as 2.9 x 10^{-9} to 3.5 x 10^{-9} $m^2 \cdot s^{-1}$, 3.7 x 10^{-9} to 4.4 x 10^{-9} $m^2 \cdot s^{-1}$ and 4.3 x 10^{-9} to 5.5 x 10^{-9} $m^2 \cdot s^{-1}$ at 40 °C, 50 °C and 60 °C, respectively. These values are within the commonly reported range for the drying of food materials, *i.e.* 10^{-9} to 10^{-11} $m^2 \cdot s^{-1}$ [35]. As expected, the moisture diffusivity was a very weak function of air velocity.

However, a slight increase of effective diffusivity was observed with increasing air velocity. On the contrary, temperature rise resulted into a more notable increase in D_{eff} values, an indication of enhanced mass transfer.
Table 4 allows a comparison between effective diffusivities and activation energies obtained in this study with those previously reported in the literature for other crops. For ground macadamia nuts, activation energies were calculated as 9.57 kJ·mol^{-1}, 19.50 kJ·mol^{-1} and 27.6 kJ·mol^{-1} at 0.7 m·s^{-1}, 1.5 m·s^{-1} and 2.5 m·s^{-1}, respectively. These activation energy values, as well, are close to those previously reported in the literature as shown in Table 4.

Table 4. *Comparison of selected effective diffusivity (D_{eff}) at 50 °C and activation energy (Ea) data from the literature with those obtained in this study*

Crop and Reference	D_{eff} **[m·s^{-2}]***	**Crop and Reference**	***Ea*** **[kJ·mol^{-1}]**
Garlic slices [6]	2 x 10^{-11}	Garlic slices [6]	17.79
Whole macadamia nuts (48 °C) [22]	2.0 x 10^{-9}	Whole macadamia nuts (48 °C) [22]	10.24
Green beans [36]	2.6 x 10^{-9}	Potato [37]	20.00
This study	3.7 x 10^{-9} to 5.5 x 10^{-9}	This study	9.57 to 27.60

A comparison was made between diffusivity and activation energy data obtained in this study with those reported for whole in-shell and out-shell macadamia nuts, using information provided in the literature [22]. Hence, on the basis of data reported in this study, it was observed that reducing the particle size of macadamia nuts resulted into shorter drying times, enhanced diffusivities and higher activation energies.

CONCLUSIONS

Bearing in mind processes such as oil extraction which require particulate matter, the drying behavior of ground macadamia nuts of particle size corresponding to 1.29 ± 0.11 mm was investigated in this study. Consistently with results previously obtained for whole macadamia nuts, Page and two-term exponential models emerged as the best semi-theoretical models to describe the thin layer drying of ground macadamia nuts. It was also found that higher temperatures and smaller granulometries would lead to shorter drying times. Effective diffusivity and activation energy were both found to increase with decreasing particle size. The data reported in this study give insights into the effect of particle size on the drying behavior of macadamia nuts. They can be used for the purpose of analyzing or designing large scale convective dryers for ground macadamia nuts.

ACKNOWLEDGEMENT

Ms. Desree Reddy's assistance during the data collection stage, while working at Mangosuthu University of Technology, is highly appreciated.

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