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Kinetics of drumstick leaves (*Moringa oleifera*) during convective drying

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Drying characteristics of drumstick leaves (*Moringa oleifera*) were investigated in a convective type dryer for a temperature range 50 to 80 C at constant air velocity, 0.5 m/s. Results indicated that drying took place in the falling rate period. The sample dried at 60 C was found better in color as compared to the samples obtained at 50, 70 and 80 C. The chroma of the samples, dried in between 50 to 80°C was ranged 4.9238 to 9.5258 and was the least, 4.9238 for the sample dried at 60 C, validating the sensory results. Moisture transfer from drumstick leaves was described by applying the Fick's diffusion model, and the effective moisture diffusivity was calculated. Effective moisture diffusivity increased with increasing temperature. An Arrhenius relation with an activation energy value of 12.50 kJ/mol expressed the effect of temperature on the moisture diffusivity. Mathematical models were fitted to the experimental data and the performance of these models was evaluated by comparing the coefficient of determination (R^2) and reduced chi-square (χ^2) between the observed and predicted moisture ratio. Verma model gave the best results for describing the drying kinetics of drumstick leaves.

Key words: Drumstick leaves, convective drying, mathematical modelling, effective moisture diffusivity, activation energy.

INTRODUCTION

Moringa oleifera commonly referred to simply as "Moringa", the most widely cultivated species of the genus *Moringa*, which is the only genus in the family *Moringaceae*. It is locally known by various names in India (Quattrocchi and Umberto, 2000) and it is considered one of the world's most useful trees, as almost every part of the *Moringa* tree can be used for

food or other beneficial applications. *Moringa* leaves are considered significant source of β -carotene, Vitamin C, protein, iron, potassium, calcium and phosphorus and are commonly dried and crushed into a powder and stored without refrigeration for months without loss of nutritional values (Fahey, 2005) and used in soups and sauces (Fuglie, 2001). *Moringa* leaves contains phytochemical, having potent anticancer and hypotensive activity and are considered full of medicinal properties and used in Siddha medicine (Rajangam et al., 2001). Various nutritional and medicinal properties of *M. oleifera* leaves have been reported by various researchers (Fahey, 2005). Drying is defined as a process of moisture removal due to simultaneous heat and mass transfer (Gogus, 1994). The drying kinetics of food is a complex phenomenon and requires dependable models to predict drying behaviour (Sharma et al., 2003). There are several studies describing the drying behaviour of various fruits, vegetables and medicinal plants such as onions (Singh and Sodhi, 2000), red pepper (Doymaz and Pala, 2002), garlic cloves (Sharma et al., 2003), ear corn (Friant et al.,

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Nomenclature: a,b,k,g,n, Empirical constants in drying models; RMSE, root mean square error; MR, moisture ratio (dimensionless); N, positive integer; M, moisture content at any time; t, drying time (min); M_e , equilibrium moisture content; L, half thickness of the slab (m); M_o , initial moisture content; E_a , activation energy (kJ/mol); R^2 , coefficient of determination; T, temperature (C); D_{eff} , effective diffusion coefficient (m^2/s); χ^2 , reduced chi-square; D_o , Arrhenius factor (m^2/s); R, gas constant, kJ/mol.K.

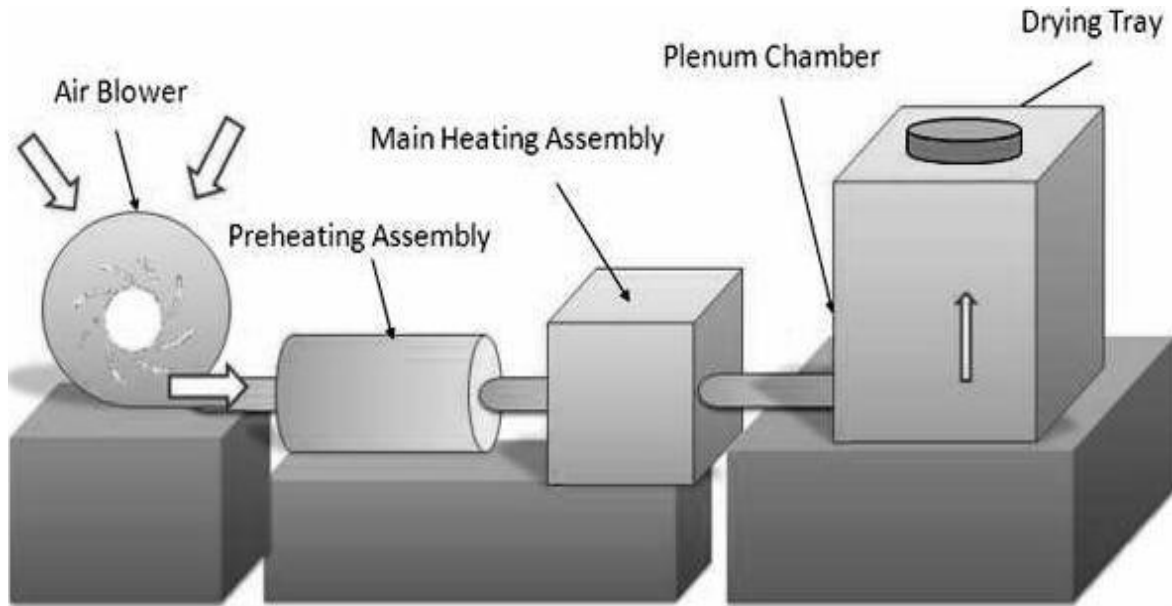


Figure1. Experimental setup.

2004), apricots (Doymaz, 2004a), mulberry (Doymaz, 2004b) and *Aloe vera* (Gulia et al., 2010).

Numbers of vegetable and medicinal plants are dried for their uses in the foods and medicines. But, the method adopted is mainly empirical in nature and requires systematic methodology for obtaining the good quality product. No systematic methodology is reported so far made for getting the product from drumstick leaves (*M. oleifera*). Therefore, the present study focuses to investigate the drying behaviour of drumstick leaves and to investigate a suitable drying model to describe the drying kinetics.

MATERIALS AND METHODS

Materials

The Drumstick leaves (*M. Oleifera*) that were used in the drying experiments were procured from region of Longowal (District Sangrur, Punjab, INDIA,). The leaves were manually separated from the stems. Initial and final moisture contents of samples were obtained by the standard method (AOAC, 1990). The initial moisture content of drumstick leaves was 276.06% (d.b.).

Drying equipment

An electric convective type dryer (Raste Enterprises, Sangli, Maharashtra, India) was modified for drying drumstick leaves, which could regulate to desired drying air temperature between 30 and 100°C with ±1°C accuracy. The dryer consisted of a preheating and heating chamber with thermostat based control unit, an electrical centrifugal fan, measurement sensors, an air-duct and a plenum chamber (Figure 1). The pre-heating control unit included an electric heater (2 kW) placed inside a duct where as the main heating control unit included 4 heaters of 1 kilowatt each. The

drying chamber was constructed from sheet iron with the cavity dimension of 420 mm × 320 mm × 400 mm. The leaves were spread in a thin layer on a steel perforated tray having opening size 1 mm × 1 mm. The drying air temperature in the chamber was measured directly using thermocouple based temperature indicator (Digital Multi-thermometer, Taiwan, Precision 1°C). The air passed from the heater at the desired temperature, entered into the chamber and mixed there and then passed through the layer of the sample. The air velocity was kept at a constant value of 0.5 m/s just above the tray surface and measured using a hot wire digital anemometer (Lutron Anemometer, Taiwan) . A digital balance with measurement precision of ± 0.1 g was used for the measurement of weight loss of the sample.

Drying experiments

Before drying experiments, initial moisture content of the examples was determined. The initial moisture content of drumstick leaves was about 74% and final moisture content of the finished product was about 3% (d.b). Four air-drying temperatures (50, 60, 70 and 80°C) were chosen to obtain the drying characteristics of drumstick leaves. Air velocity was set at 0.5 m/s. The sample size was kept constant at 50 g for each run. After the dryer reached steady-state conditions for the set points (at least 1 h), the drumstick leaves (sample thickness: 1.0 ± 0.1 cm) were distributed uniformly into the tray. Moisture loss was recorded at every 5 min intervals during drying. Drying was continued until the moisture content of sample reached about 3% (d.b). The drying was continued till weight became constant. The experiments were conducted in duplicate.

Mathematical modelling

The moisture ratio (MR) and drying rate during drying experiments were calculated using the following equations:

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

Table 1. List of models with references.

Model name	Model	References
Newton	MR = exp(-k t)	Mujumdar (1987)
Page	MR = exp(-k t ⁿ)	Diamante and Munro (1993)
Wang and Singh	MR = 1 + at + bt ²	Wang and Singh (1978)
Logarithmic	MR = a exp(-k t) + c	Yagcioglu et al. (1999)
Two-term exponential	MR = a exp(-k t) + (1-a)exp(-k a t)	Sharaf-Eldeen et al. (1980)
Handerson and Pabis	MR = a exp(-k t)	Henderson and Pabis (1961)
Verma et al.	MR = a exp(-k t) + (1-a)exp(-g t)	Verma et al. (1985)
Magee	MR = a + kt ^{1/2}	Magee et al. (1983)

$$\text{Drying Rate} = \frac{M_t - M_{t+dt} - M_e}{dt} \quad \dots (2)$$

where, MR, M₀, M_e, M_t and M_{t+dt} are the moisture ratio, initial moisture content, equilibrium moisture content, moisture content at t and moisture content at t + dt (kg moisture/kg dry matter), respectively, t is drying time (min).

The air-drying curves were fitted with different mathematical models as given in Table 1. The regression analysis was performed using the STATISTICA computer program. Non-linear regression, which used to evaluate goodness of fit of the mathematical models to the experimental data are coefficient of determination (R²) and the reduced chi -square (χ²), was used for data analysis. The higher value for R² and the lower values for χ² and root mean square error analysis (RMSE) indicate the better fitness of model (Sarsavadia et al., 1999; Togrul and Pehlivan, 2003). These parameters were calculated as follows:

$$\chi^2 = \frac{\sum_{i=1}^n (MR_{exp,i} - MR_{pre,i})^2}{N - n} \quad \dots (3)$$

$$RMSE = \left[\frac{1}{N} \sum_{i=1}^n (MR_{exp,i} - MR_{pre,i})^2 \right]^{1/2} \quad \dots (4)$$

where, MR_{exp, i} is the ith experimentally observed moisture ratio, MR_{pre, i} the ith predicted moisture ratio, N is the number of observations and n is the number constants.

Color evaluation of *M. oleifera* leaves

Color of the samples was evaluated in terms of L, a, b, E, H by using colorimeter (Model: i5 – 150133, Gretag Macbeth, UK). Sensory evaluation was also carried out for the color by the ranking method (Ranganna, 1986).

RESULTS AND DISCUSSION

Drying characteristics of drumstick

Moisture ratio and drying rate of the samples were calculated using Equations (1) and (2). Figure 2 shows

the drying curves of drumstick leaves at different air temperatures. The drying rate decreased continuously throughout the drying period. It is obvious from Figure 2 that the constant rate period was absent and the drying process of drumstick leaves took place in falling rate period. These results are in good agreement as compared to the earlier studies on herbal leaves by Doymaz et al. (2006). The drying times to reach the equilibrium moisture content for the fresh sample were 600, 480, 410 and 390 min at 50, 60, 70 and 80°C, respectively. As the temperature was increased by difference of 10°C, from 50 to 80°C the drying time decreased by 20, 14.58 and 5.13%, correspondingly. Maximum reduction of drying time was obtained when the drying temperature was increased from 50 to 60°C as compared from 60 to 70°C and 70 to 80°C, respectively. The variations of drying rate and moisture ratio with drying time at temperatures of 50, 60, 70 and 80°C are given in Figures 3 and 4 respectively. The moisture ratio reduced exponentially as the drying time increased (Doymaz, 2007). Continuous decrease in moisture ratio indicates that diffusion has governed the internal mass transfer. A higher drying air temperature decreased the moisture ratio faster due to the increase in air heat supply rate to the leaves and the acceleration of moisture migration (Demir et al., 2004). Experimental results showed that drying air temperature is effective parameter for the drying of drumstick leaves.

The drying process took place in a falling rate period except a very short accelerating period in the beginning (Figure 5). It can be seen that at higher moisture content, the increase in temperature has more considerable effect on the drying rates as compared to lower temperatures, which is almost negligible towards the end. It was further observed that the drying rate or moisture loss was faster at the beginning than that at the end. The reduction in the drying rate at the end of drying may be due to the reduction in moisture content as drying advances (Sharma and Prasad, 2001). Thus, a higher drying air temperature produced a higher drying rate and consequently the moisture ratio decreased (Lahsasni et al., 2003) (Figure 6). The dried samples obtained at 50, 60, 70 and 80°C were evaluated for sensory parameter

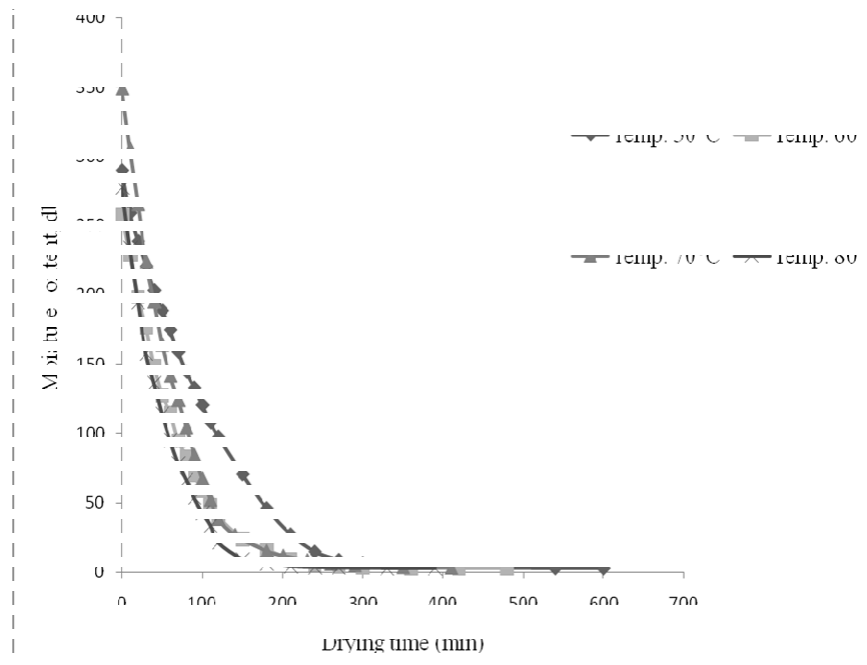


Figure 2. Drying curves of drumstick leaves at selected temperatures.

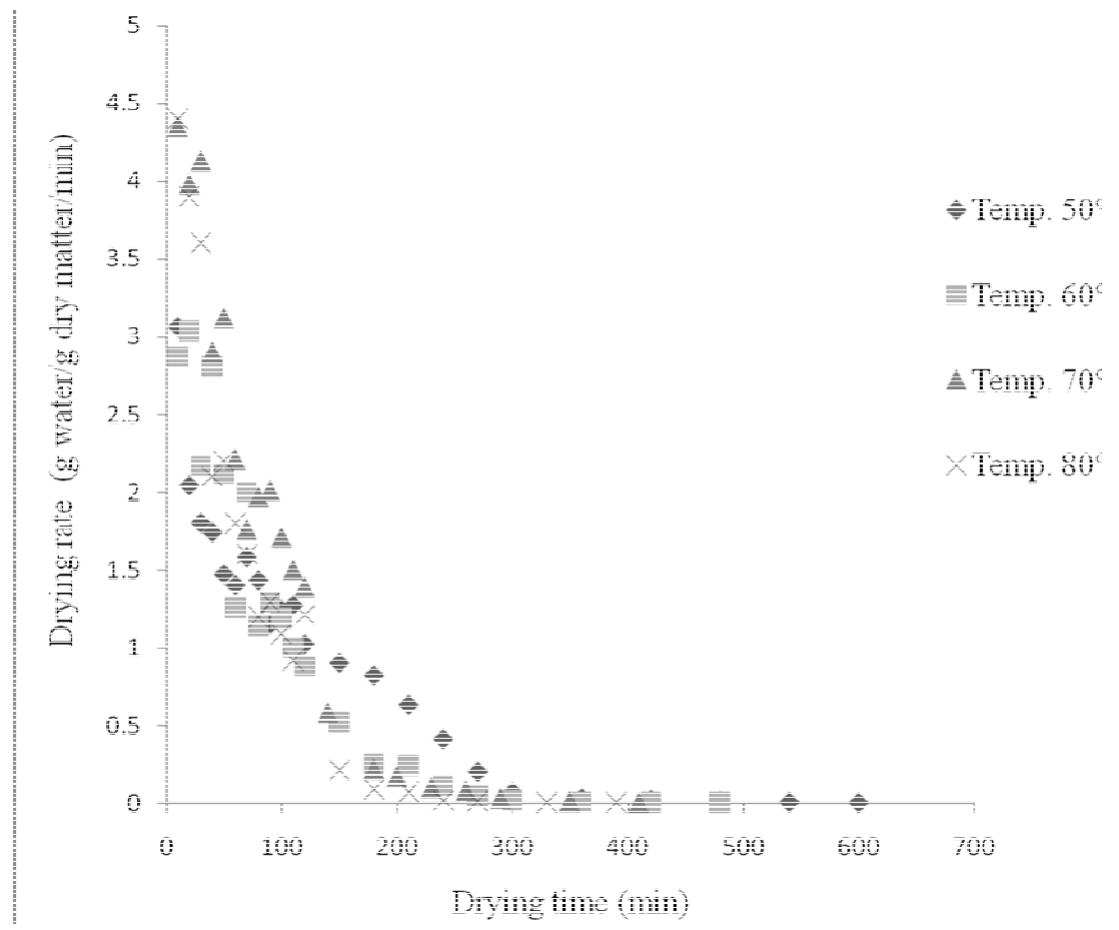


Figure 3. Drying rate versus drying time of drumstick leaves at different temperatures.

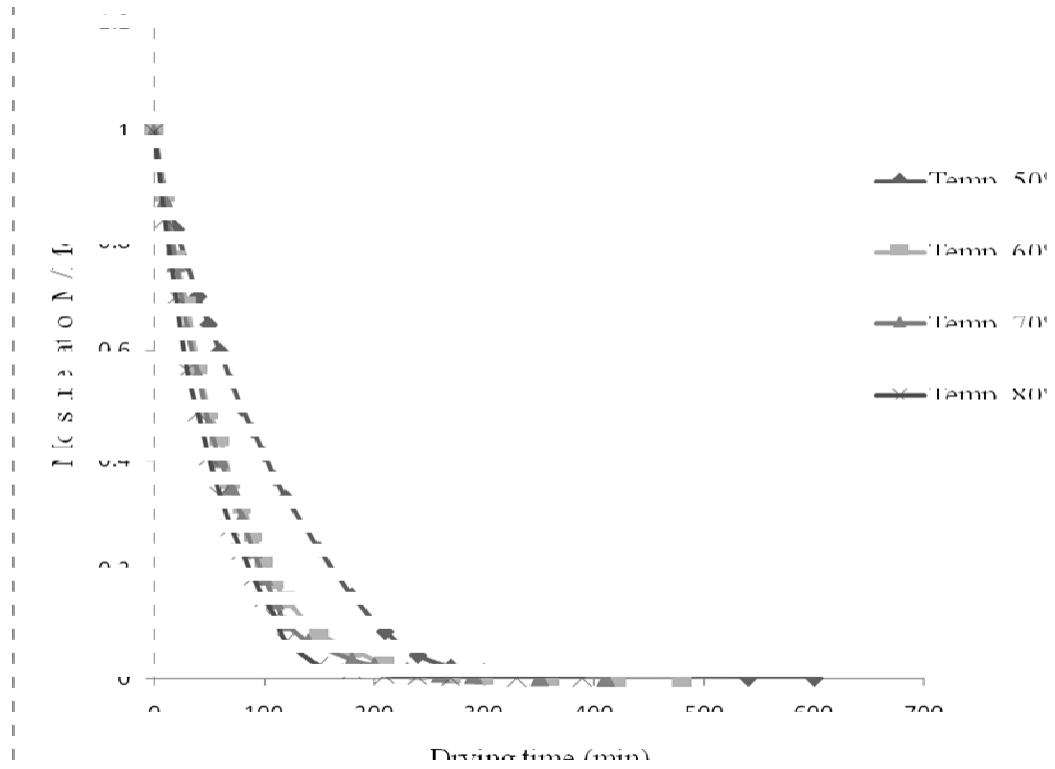


Figure 4. Moisture ratios versus drying time of drumstick leaves at different temperatures.

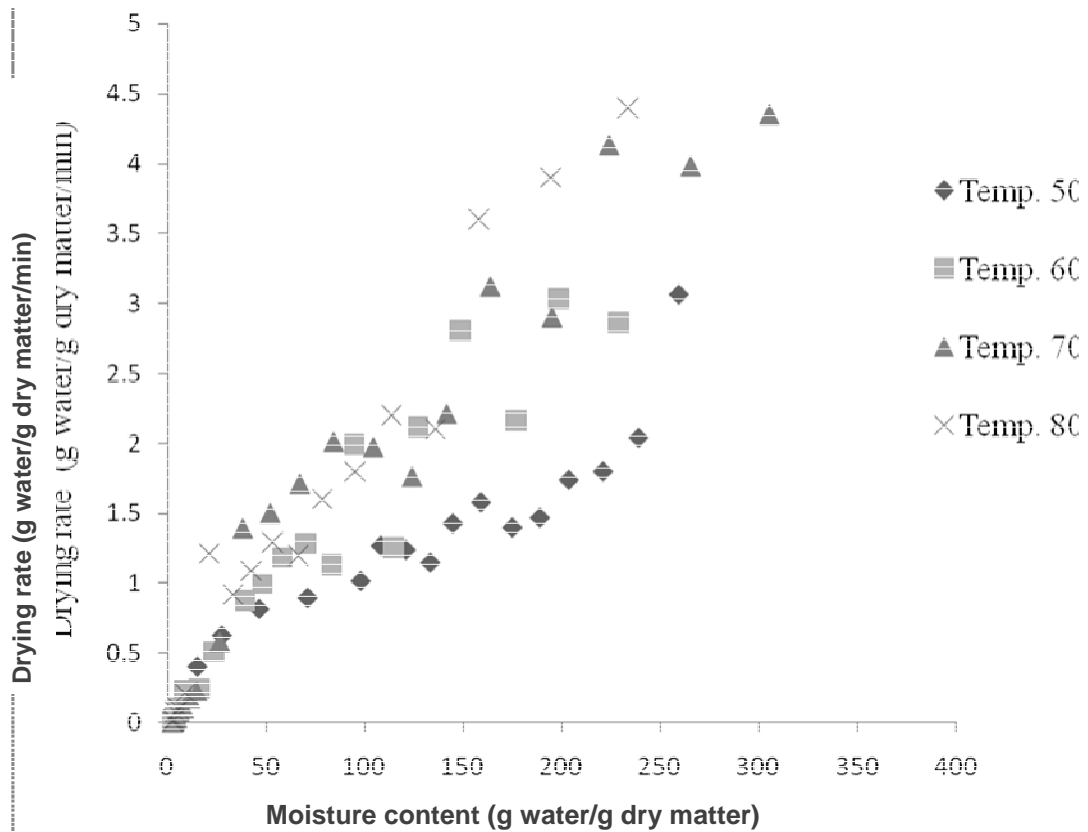


Figure 5. Drying rate versus moisture content of drumstick leaves at different temperatures.

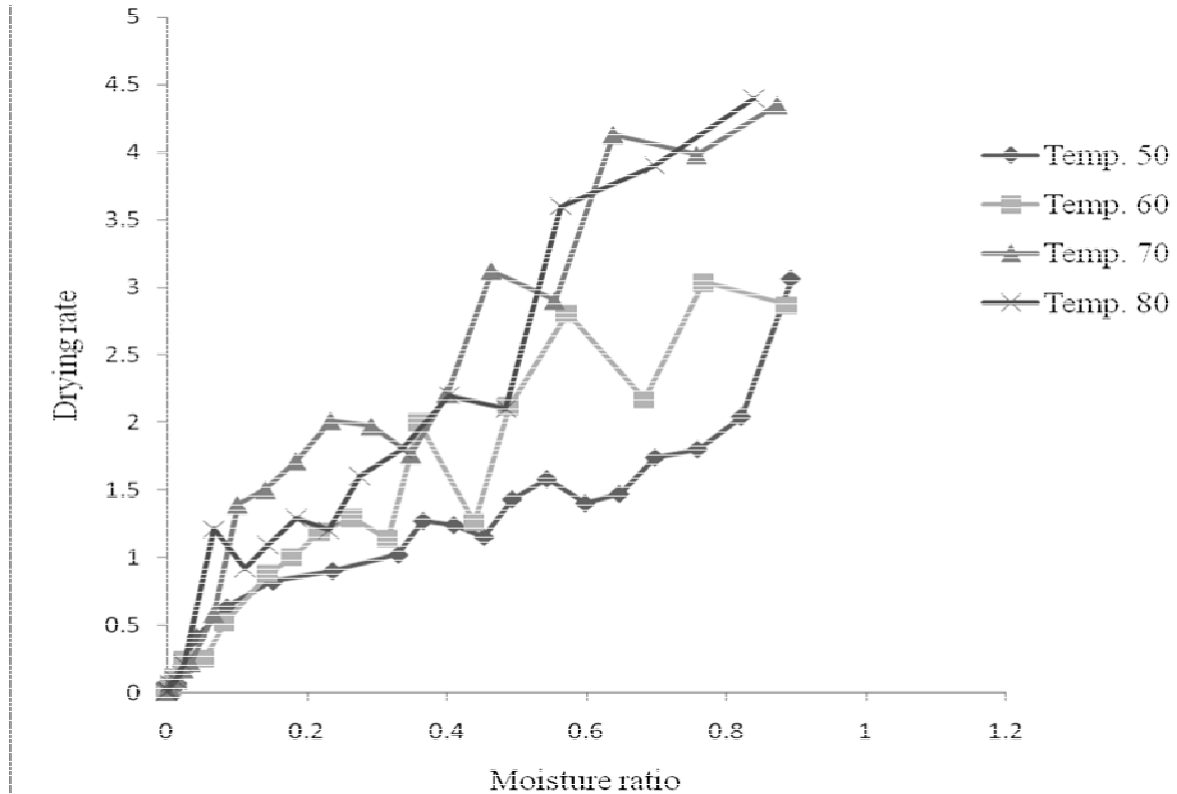


Figure 6. Variation of drying rate with temperature during drying of drumstick leaves.

Table 2. The L*, a*, b*, E* values and hue angle of dried drumstick leaves samples.

Temperature (°C)	L*	a*	b*	E*	h*
Control	48.674	-3.121	5.349	-	-59.7375
50	49.020	-2.963	11.346	6.0091	-75.3641
60	49.957	-2.700	10.084	4.9238	-75.0106
70	42.044	3.059	8.280	9.5258	69.7235
80	43.878	-0.510	8.799	6.4592	-86.6828

and color. The sample dried at 60°C was found better as compared to the samples obtained at 50, 70 and 80°C because at 50°C retention time was more and at 70 and 80°C retention time was less but color degradation was at faster rate. The samples dried at 50 to 80°C were not liked by the sensory panelists. The color of the samples was also examined by the colorimeter as shown in the Table 2. The sample dried at 50 and 60°C were found closer to control samples. L, a, b values for control and dried samples at 50 and 60 were 48.674, -3.121, 5.349 and 49.020, -2.963, 11.346 and 49.957, -2.7 and 10.084, respectively. The chroma of the samples, dried in between 50 to 80 was ranged 4.9238 to 9.5258 and was the least, 4.9238 for the sample dried at 60, validating the sensory results (Table 2).

Mathematical models for fitting of drying curves

The moisture ratio data of drumstick leaves dried at different temperatures were fitted into the drying models (Table 1). The coefficient of correlation and results of statistical analyses are listed in Table 2. In all cases, the R^2 values for the mathematical models were greater than 0.90, except the Magee model indicating the fitness of the models in predicting the data. The results showed that highest values of R^2 and lowest values of $RMSE$ were obtained with the Verma model (Table 3). Therefore, this model can be considered to represent the drying behaviour of drumstick leaves in a convective type dryer. Demir et al. (2004), have reported similar results for air drying of bay leaves. The variation of the Verma

Table 3. The fitness of different models at different temperature.

Model	Temperature (T), °C	Coefficient	Coefficient of determination (R^2)	chi-square (χ^2)	RMSE
Newton	50	k:0.009448	0.9963	0.000808	0.000387
	60	k:0.014621	0.9983	0.000335	0.00016
	70	k:0.015982	0.9975	0.000463	0.000222
	80	k:0.019028	0.9980	0.000350	0.000167
Page	50	k: 0.005126, n:1.132298	0.9980	0.000448	0.000205
	60	k: 0.008585, n:1.124476	0.9998	4.98E-05	2.26E-05
	70	k:0.008748, n:1.141749	0.9994	0.000123	5.65E-05
	80	k:0.013261, n:1.086825	0.9989	2.11E-04	9.61E-05
Wang and Singh	50	a: -0.005812, b:0.000007	0.9658	0.007623	0.003494
	60	a:-0.007800, b:0.000013	0.9129	0.017419	0.007918
	70	a:-0.008758, b:0.000017	0.9317	0.012987	0.005952
	80	a:-0.009728, b:0.000020	0.9035	0.017134	0.007788
Logarithmic	50	a:1.041994, k:0.008901, c:-0.034708	0.9976	0.000566	0.000247
	60	a:1.040803, k:0.014573, c:-0.014792	0.9991	0.000195	8.40E-05
	70	a: 1.042546, k:0.015910, c:-0.014653	0.9984	0.000323	0.000142
	80	a:1.029633, k:0.018388, c:-0.018718	0.9987	0.000247	0.00010
Two-term exponential	50	a:0.002742, k:3.428842	0.9962	0.00085	0.000394
	60	a:0.002708, k:5.378195	0.9982	0.00037	0.000168
	70	a:0.002679, k:5.944404	0.9974	0.000501	0.00023
	80	a:0.003650, k:5.190622	0.9980	0.000381	0.000173
Handerson and Pabis	50	a:1.017663, k:0.009655	0.9964	0.000804	0.000369
	60	a:1.031013, k:0.015121	0.9988	0.000249	1.13E-04
	70	a:1.033363, k:0.016535	0.9981	0.00038	0.000174
	80	a:1.018206, k:0.019368	0.9982	0.000336	0.000153
Verma et al.	50	a:-11.1110, k:0.0153, g:0.0147	0.9984	0.000374	0.000164
	60	a:3.444414, k:0.021023, g:0.024878	0.9998	4.33E-05	1.87E-05
	70	a:4.929988, k:0.023779, g:0.026732	0.9995	0.000109	4.78E-05
	80	a:5.315766, k:0.026105, g:0.028367	0.9990	0.000192	8.29E-05
Magee	50	a:0.655010, k:-0.003150	0.8450	0.032405	0.014852
	60	a:0.573451, k:-0.003602	0.7863	0.039898	1.81E-02
	70	a:0.559793, k:-0.003908	0.7902	0.036969	0.016944
	80	a:0.523483, k-0.004283	0.7527	0.040437	0.01838

model was established by comparing the experimental data with predicted values for each drying curve. The plotted responses in Figure 7 demonstrates that the data points follows a straight line at 45° angle signifying the suitability of the model in describing the drying of the drumstick leaves.

Effective moisture diffusivity and activation energy

The results obtained have shown that internal mass transfer resistance due to presence of falling rate drying period controls drying time. Therefore, experimental results can be interpreted by using Fick's diffusion equation.

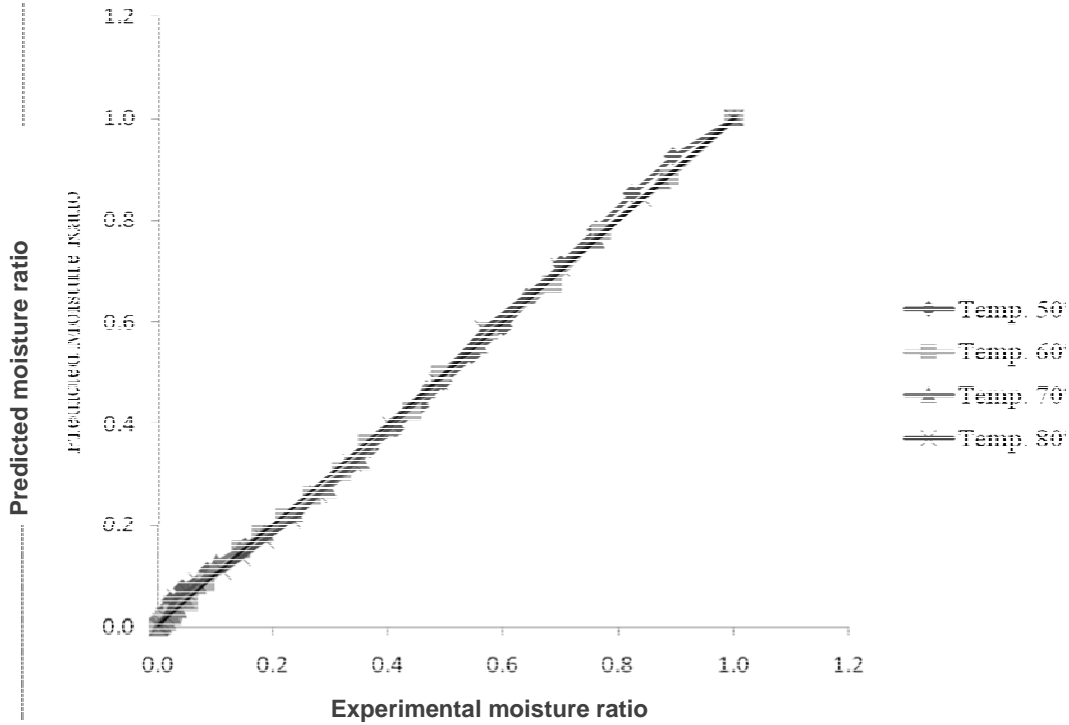


Figure 7. Experimental vs. predicted values using the Verma model for different air temperatures.

Table 4. Calculated effective moisture diffusivity at different drying temperatures.

S/No.	Temperature (°C)	Effective diffusivity ($D_0 \times 10^{-9} \text{ m}^2/\text{sec}$)
1.	50	2.40
2.	60	3.04
3.	70	3.21
4.	80	3.89

The effective diffusivity D_{eff} is determined by using the analytical solutions of Fick's second law Crank (1975), represented as:

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

Where, D_{eff} is the effective moisture diffusivity (m^2/s), L is the half thickness of the slab in samples (m), and n is positive integer. Simplifying by taking the first term of the series solution:

$$MR = \frac{8}{\pi^2} \exp\left(-\frac{\pi^2 D_{\text{eff}} t}{4L^2}\right) \quad \dots (6)$$

Effective moisture diffusivity is also typically calculated by using slope of Equation (6), namely, when natural logarithm of MR versus time was plotted, straight line with

a slope was obtained:

$$\text{Slope} = \frac{-\pi^2 D_{\text{eff}}}{4L^2} \quad \dots (7)$$

The calculated values of D_{eff} for 50, 60, 70 and 80 °C were 2.40, 3.04, 3.21 and 3.89 $\times 10^{-9} \text{ m}^2/\text{s}$ (Table 4). The values lie in the general range of 10^{-11} to $10^{-9} \text{ m}^2/\text{s}$ for food materials (Zogzas et al., 1996). It can be seen that the values of D_{eff} increased with increasing temperature. As the hypothesis was presumed that the diffusion has governed the internal mass transfer and mass transfer rate was highest at 80 C, which also verified from the above values of effective moisture diffusivity. The highest effective moisture diffusivity value is 3.89 $\times 10^{-9} \text{ m}^2/\text{s}$ at 80 C. An Arrhenius type of equation is generally used to model the effect of temperature on the effective moisture diffusivity as follows:

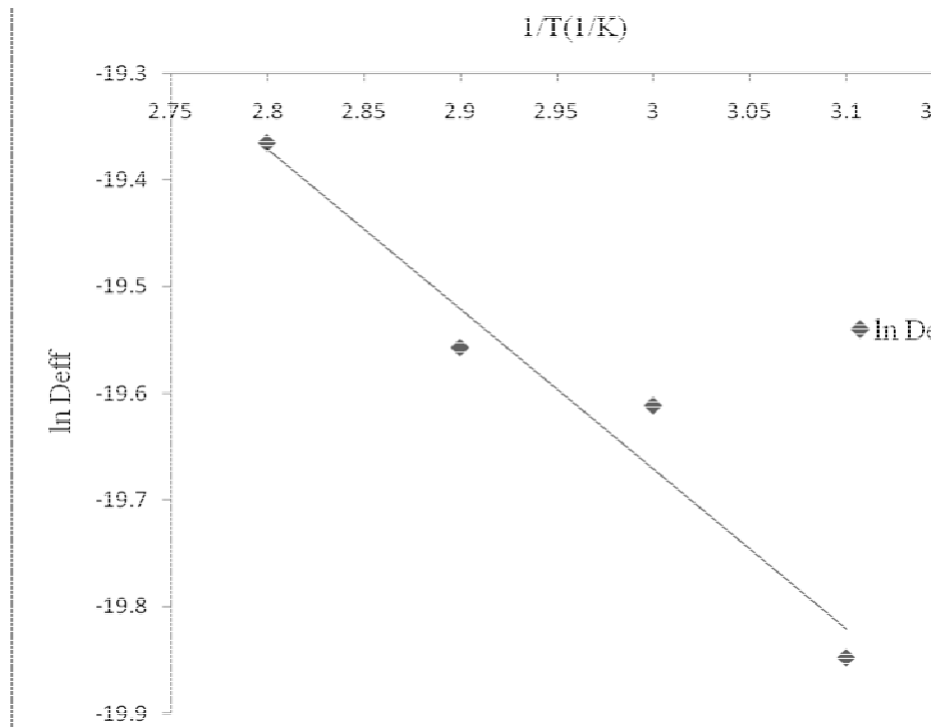


Figure 8. The Arrhenius type relationships between effective diffusivity and drying temperature.

$$D_{eff} = D_0 \exp\left(-\frac{E_a}{R(T+273.15)}\right) \quad \dots\dots\dots (8)$$

where, D_0 is the Arrhenius factor (m^2/s), E_a is the activation energy for the moisture diffusion (kJ/mol), R is the universal gas constant (kJ/mol K) and T is drying air temperature (C).

The energy level of the molecule to initiate a chemical reaction is generally represented as activation energy. According to Equation (8), straight line was obtained when the temperature influence on D_{eff} was evaluated (Figure 8). The activation energy for drumstick leaves was evaluated as 12.50 kJ/mol. These results are in coherence with the findings of Ibrahim et al. (2006) who reported activation energy for dill and parsley leaves.

Conclusions

M. oleifera leaves were dried at 50, 60, 70 and 80°C temperature, the drying rate decreased continuously throughout the drying period. Constant rate period was absent and the drying process of drumstick leaves took place in falling rate period. Drying time decreased considerably with increased temperature. Verma model was the best among the selected models for describing the drying behaviour of drumstick leaves. The effective moisture diffusivity ranged from 2.40×10^{-9} to 3.89×10^{-9}

m^2/s , when dried at 50 to 80°C. The activation energy was calculated as 12.50 kJ/mol for drumstick leaves. Dried leaves when evaluated on the basis of color by sensory, L , a , b values and drying time, the temperature, and 60°C was found to be the optimum in terms of energy efficiency and product quality.

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