

Mathematical modeling of thin layer drying kinetics and moisture diffusivity study of elephant apple

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Abstract

In the present work, drying kinetics of elephant apple (*Dillenia indica*) was studied at four different drying temperatures i.e. 50, 60, 70 and 80°C using a laboratory scale tray drier. A 4 mm thick uniform layer of the fruit pulp was dried at the above mentioned temperatures and the subsequent changes in weights were noted at intervals of 30 minutes until a constant weight was achieved. The drying rates gradually increased with increasing drying temperatures. Drying took place during the falling rate period which was evident from the experimental drying curves. The drying data was applied and fitted to 14 different thin-layer drying models available in the literature. Amidst all the models, the two term exponential model gave the best fit with the highest determination coefficient, R^2 (0.998) and the lowest chi-square χ^2 (2.262×10^{-4}) values. A knowledge of effective moisture diffusivity helps to design and model various mass transfer processes like dehydration, adsorption and desorption of moisture during storage. Fick's law was used to determine effective moisture diffusivity and the later had a linear relationship with temperature. Its value ranged from 1.095×10^{-10} - 2.283×10^{-10} m²/s within the experimental temperature range of 50-80°C. The effective moisture diffusivity was correlated with temperature using Arrhenius equation. The activation energy, an indicator of the energy required for moisture removal from a solid matrix was found to be 21.95kJ/mol.

Keywords

Elephant apple

Drying kinetics

Mathematical model

Activation energy

Effective moisture diffusivity

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Introduction

Elephant apple (*Dillenia indica*) is consumed both in fresh and in cooked form in curries and has been traditionally used for making jams, jellies, pickles, chutneys and juices. Besides its culinary purpose, the fruit also has promising health benefits. The fruit is indigenously used in Ayurveda to treat nervousness, abdominal distress and fatigue (Janick *et al.*, 2008). The juice of fruit is used as cooling beverage in fever, diarrhea, and dysentery. Literature reviews have revealed that the plant has great medicinal value including antimicrobial (Nazma *et al.*, 2009), antioxidant (Deepa *et al.*, 2011), analgesic (Badrul *et al.*, 2012), anti-inflammatory (Yeshwante, 2009), dysentery (Das *et al.*, 2008), antidiabetic (Kumar *et al.*, 2011) activities. The fruits and the juice of the plant are traditionally used for the treatment of various diseases including diabetes mellitus (Talukdar *et al.*, 2012). The fruit's fleshy sepals are rich in tannins, malic acid, arabinogalactan and glucose. They also contain betulin, betulinic acid and flavonoids (Talukdar *et al.*, 2012).

The availability of elephant apple is limited, as the fruits ripen only during October-January. This necessitates the use of drying to preserve the fruits for year round use. Drying is the most widely employed

method for preserving food materials; which is based on reduction of the water activity values through moisture removal to achieve physicochemical and microbiological stability (Moreira *et al.*, 2008). Drying allows safe storage over an extended period and also, brings about substantial reduction in weight and volume, minimizing packaging, storage and transportation costs.

Mathematical modeling of the drying processes and equipment is the most important aspect of drying technology. Its purpose is to allow design engineers to choose the most suitable operating conditions and then size the drying equipment and drying chamber accordingly to meet desired operating conditions. There are many empirical or semi-empirical models for the simulation of drying process (Vega-Galvez *et al.*, 2010) like Page model, Modified Page, Two term, Logarithmic etc.

A knowledge of Effective moisture diffusivity is necessary for designing and modeling mass transfer processes such as dehydration, adsorption and desorption of moisture during storage. The effective diffusivity depends on the drying air conditions besides the variety and composition of the material (Rizvi, 1986). The relationship between effective diffusivity and absolute temperature generally follows a first order rate process described by Arrhenius equation

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and the relationship can be generally characterized by Activation energy. The activation energy serves as an indication of the minimum amount of energy required to initiate moisture diffusion from a solid matrix. The knowledge of effective moisture diffusivity and activation energy is necessary for designing and modelling the mass transfer processes such as dehydration or moisture adsorption during storage. Literature survey has revealed that there is a dearth of substantial research on the drying behavior of elephant apple. Consequently, the present study has been done to examine the thin layer drying of elephant apple pulp at different air drying temperatures. The drying kinetics of the fruit was modeled using different drying models. Moisture diffusivity study was carried out using the Arrhenius equation and the energy of activation was estimated.

Materials and Methods

Raw material

Dillenia indica (elephant apple) has originated from Indonesia and belongs to family Dilleniaceae. Fresh fruits were procured from the local market (Tezpur, Assam). The fruits were washed thoroughly and cut into small fractions. The edible portion of the fruit is the gelatinous flesh surrounding the pistons and the crunchy petals. The edible portion was pulped in a mixer grinder. Pulped samples were spread at a uniform thickness (4 mm) to allow maximum surface exposure to convective hot air. The samples were then dried in a laboratory scale tray drier (Model No.IK-112, IKON Instruments, and New Delhi) at constant temperatures of 50, 60, 70 and 80°C.

Experimental procedure

In order to study the influence of temperature on drying kinetics, experiments were conducted at four different drying temperatures which are 50, 60, 70 and 80°C. Mature fruits were procured and thoroughly washed and cleaned. A uniform fruit pulp was obtained using a mechanical grinder. The resulting pulp was spread in uniform thin layers of approximately 2 mm thickness which allowed sufficient exposure to drying environment (Akpınar and Bicer, 2008). The sample weights were noted after every 30 minutes. The drying was continued till equilibrium moisture content was achieved by the sample. The moisture content was calculated using the oven drying method (Ranganna, 1986). 5 g sample was kept in the oven and dried at 105°C for 24 hours.

Modelling of drying kinetics

As the drying advances, the sample moisture

content eventually decreases. During falling rate period, moisture removal is triggered by diffusion of moisture from inside towards the surface. This consequently results in mass transfer of the moisture via evaporation to the surrounding environment of the product. This mechanism of diffusion is described by Fick's second law of diffusion as follows.

The moisture ratio (MR) during drying experiments is presented by Eq.(1)

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

Where M_t , M_o , and M_e relate to the moisture content at any drying time, initial moisture content and equilibrium moisture content (kg water/kg dry matter), respectively.

Various semi empirical drying models of the lumped parameter type like the Newton's model have been described in literature for studying the thin layer drying of food samples. For drying model selection, moisture ratios were fitted to 14 well known thin layer drying models which are generally applicable for drying of fruits and vegetables. The determination coefficient (R^2) and the reduced chi-square, (χ^2) were considered as the primary criteria for selecting the best equation to describe the drying process. This provides the description how experimental values are deviated from the predicted values for a particular model.

The coefficient of determination (R^2) and Chi-square (χ^2) values were evaluated by application of Equations (2) and (3) respectively.

$$R^2 = 1 - \frac{SS_{res}}{SS_{tot}} \quad (2)$$

$$\chi^2 = \frac{\sum (O-E)^2}{E} \quad (3)$$

Where, SS_{res} = residual sum of square; SS_{tot} = total sum of square; O=observed value; E=Expected value.

The highest values of the R^2 and lowest values of the χ^2 , determine the goodness of the fit (Goyal *et al.*, 2006; Khawas *et al.*, 2015).

Calculation of effective moisture diffusivity and activation energy

Drying of food materials is a function of internal diffusion and generally occurs in the falling rate period. Several mathematical models have been proposed using Fick's second law to describe drying processes during falling rate period. The Fick's second law of diffusion is presented in Eq. (4).

$$\delta M / \delta t = D \delta^2 M / \delta x^2 \quad (4)$$

Where, D = diffusivity (m²/sec); δM/δt = moisture content (db) per unit time (sec) and x = thickness (m).

By considering the geometry as a slab having moisture distributed uniformly at a concentration M₀ and diffusion taking place only in the X direction, on the basis of the assumptions stated below, the mathematical Eq. 5-8 could be deduced.

Assumptions

- (1) Moisture is initially distributed uniformly throughout the mass of a sample.
- (2) Mass transfer is symmetric with respect to the center.
- (3) Surface moisture content of the sample instantaneously reaches equilibrium with the condition of surrounding air.
- (4) Resistance to the mass transfer at the surface is negligible compared to internal resistance of the sample.
- (5) Mass transfer is by diffusion only.
- (6) Diffusion coefficient is constant and shrinkage is negligible

Initial conditions (t = 0):

$$M = M_0 \quad 0 \leq X < L \tag{5}$$

Boundary conditions (t > 0):

$$\left. \frac{\partial M}{\partial X} \right|_{x=0} = 0 \tag{6}$$

$$M = 0 \quad X = L \tag{7}$$

By solving Eq. (4), considering the conditions expressed in Eq. (5) - (7), unsteady state diffusion equation for slab geometry can be presented as shown in Eq.(8) (Crank, 1975).

$$MR = \frac{(M_t - M_e)}{(M_0 - M_e)} = \frac{8}{\pi^2} \sum_{n=0}^{\infty} \frac{1}{(2n+1)^2} \exp \left[\frac{-(2n+1)^2 \pi^2 D_e t}{4L^2} \right] \tag{8}$$

Where MR is moisture ratio, n =the number of terms taken into consideration, t = time of drying in seconds, D_e = effective moisture diffusivity in m²/s and L = half the thickness of slab (m).

Neglecting higher order terms and considering n=0, Eq. (8) reduces to Eq. (9)

$$MR = \frac{8}{\pi^2} \exp \left[\frac{-\pi^2 D_e t}{4L^2} \right] \tag{9}$$

Taking logarithm on both sides of Eq. (9) results in Eq. (10).

$$\ln MR = \left(\ln \frac{8}{\pi^2} \right) - \frac{\pi^2 D_e t}{4L^2} \tag{10}$$

The diffusion coefficient is calculated by the method of slopes. From the slope of the plot of ln MR versus time at different temperatures effective

moisture diffusivity ‘De’ is calculated by application of Eq. (11).

$$\text{slope} = - \frac{\pi^2 D_e}{4L^2} \tag{11}$$

The energy of activation was calculated using an Arrhenius type equation (Lopez *et al.*, 2000) as shown in Eq. (12).

$$D_e = D_0 \exp \left(- \frac{E_a}{RT} \right) \tag{12}$$

Where E_a is the energy of activation (kJ/mol), R is universal gas constant (8.3143 kJ/mol), T is absolute air temperature (K), and D₀ is the pre-exponential factor of the Arrhenius equation (m²/s). The activation energy was calculated from the slope of the Arrhenius plot (Eq. 13), i.e. lnD_e versus 1/T

$$\ln D_e = \ln D_0 - \frac{E_a}{RT} \tag{13}$$

From Eq. (13), a plot of ln D_e versus 1/T gives a slope (m) given by Eq. (14)

$$m = - \frac{E_a}{R} \tag{14}$$

The drying process was discontinued once no further reduction in weights occurred. Moisture content data were converted to moisture ratio and then fitted to 14 different thin layer drying models listed in Table 1.

Results and Discussions

The fruit pulp was dried at 50, 60, 70 and 80°C till it attained equilibrium moisture content. The drying of the fruit followed falling rate period of drying. Moisture ratio was computed using the moisture content data of the samples obtained at different time intervals at different drying temperatures. The variation of moisture ratio with time at different drying temperatures has been depicted in Figure 1. The moisture content decreased exponentially with elapsed drying time under different drying temperatures. As the drying temperature increased at the specified thickness of the product the drying curve exhibited a steeper slope for all convective drying conditions, implying that drying rate increased with increase in drying air temperature within the experimental limits. This resulted in substantial decrease in drying time when higher air temperature was used. The influence of the temperature on the drying rate is also dependent on type of the food product. From the drying curves (Figure 1), it was observed that the value of moisture ratio decreases rapidly with

Table 1. Drying kinetic models considered for thin layer drying of elephant apple

Model no.	Model name	Model	References
1.	Newton	$MR = \exp(-kt)$	Westerman <i>et al.</i> , 1973
2.	Page	$MR = \exp(-kt^n)$	Page, 1949
3.	Modified page	$MR = \exp[-(kt)^n]$	Yaldiz <i>et al.</i> , 2001
4.	Henderson and Pabis	$MR = a \exp(-kt)$	Yagcioglu <i>et al.</i> , 1999
5.	Logarithmic	$MR = a \exp(-kt) + c$	Yaldiz and Ertekin, 2001
6.	Two term	$MR = a \exp(-k_0 t) + b \exp(k_1 t)$	Rahman <i>et al.</i> , 1998
7.	Two term exponential	$MR = a \exp(-kt) + (1 - a) \exp(-kt)$	Yaldiz <i>et al.</i> , 2001
8.	Wang and Singh	$MR = Mo + at + bt^2$	Ozdemir and Devres, 1999
9.	Approximation of diffusion	$MR = a \exp(-kt) + (1 - a) \exp(-k_b t)$	Yaldiz and Ertekin, 2001
10.	Verma <i>et al.</i>	$MR = a \exp(-kt) + (1 - a) \exp(-gt)$	Verma <i>et al.</i> , 1985
11.	Modified Henderson and Pabis	$MR = a \exp(-kt) + b \exp(-gt) + c \exp(-h)$	Karathanos, 1999
12.	Aghabashlo model	$MR = \exp\left[-\frac{k_1 t}{(1 + k_2 t)}\right]$	Aghabashlo <i>et al.</i> , 2008
13.	Weibull	$MR = \exp\left(-\frac{t}{a}\right)^b$	Corzo <i>et al.</i> , 2008
14.	Midilli <i>et al.</i>	$MR = a \exp(-kt^n) + bt$	Midilli <i>et al.</i> , 2002

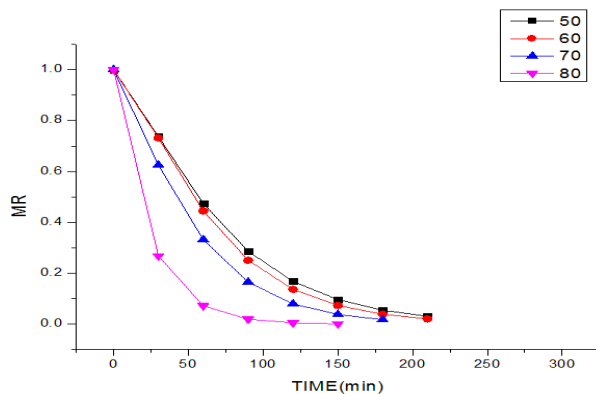


Figure 1. Plot of MR versus time at four different drying temperatures

increase of drying temperature due to increase in drying rate. The moisture ratio also decreased with progressing drying time. Similar findings have been reported by other researchers (Togrul *et al.*, 2003; Hii *et al.*, 2009; Toyosi *et al.*, 2011). Higher drying air temperatures resulted in an increase of the drying rate. The increase in the temperature reduces the relative humidity of drying air and hence increases the moisture gradient between the product and the air surrounding the food product. Higher temperature increases the water mobility inside the product and also increases the energy available in the medium for water evaporation to facilitate the drying process. In the initial stage of drying, the drying rates were found to be faster due to availability of high amount of free moisture, which was easily removed. As the drying

progressed, the drying rates increasingly decreased with time.

Mathematical modelling

The moisture ratio data was fitted to 14 different semi-empirical models as reported in Table 1. The results (R^2 and χ^2 values) obtained on fitting the experimental data to the thin layer drying models are represented by Table 2. The models were analyzed based on the statistical parameters, determination coefficient (R^2) and reduced chi-square (χ^2) values. The best model on the grounds of highest R^2 and lowest χ^2 value was found to be two term exponential model. Similar basis was used for drying model selection of pumpkin (Guine *et al.*, 2011, carrots (Doymaz, 2004a), green chili (Ahmed and Shivhare, 2001).

The selected model can be described by the following set of Eqs.15-17.

$$MR = a \exp(-kt) + (1 - a) \exp(-kat) \quad (15)$$

The parameters 'a' and 'k' are function of temperature and can be represented in terms of first order kinetic rate equation as shown in Eq. 16-17.

$$k = 0.0034 \times \exp(0.033T) \quad R^2 = 0.961 \quad (16)$$

$$a = 1.3872 \times \exp(0.0047T) \quad R^2 = 0.958 \quad (17)$$

Table 2. Model constants along with statistical parameters at different drying temperatures

Model no.	Model name	Average values of model constants	R ² value	χ ² value
1.	Newton	k=0.0146	0.927	0.00182
2.	Page	k=0.0041, n=1.2881	0.916	0.00051
3.	Modified page	k = 0.0140, n = 1.2835	0.979	0.00051
4.	Henderson and Pabis	a = 1.0199, k= 0.0189	0.936	0.02501
5.	Logarithmic	k= 0.0147, a = 1.1197, c= -0.1146	0.921	0.02830
6.	Two term	a = 2.61165, b = -1.61165 k ₀ = 0.07301, k ₁ = 205.09803	0.972	0.00083
7.	Two term exponential	a=1.884, k=0.03032	0.997	0.00023
8.	Wang and Singh	M ₀ =0.91091, a=-0.00754, b=1.45943×10 ⁻⁵	0.949	0.00670
9.	Approximation of diffusion	a=-112.11125, b=0.99334, k=0.02578	0.925	0.00040
10.	Verma <i>et al.</i>	a = -0.22422, k = 1.95255, g= 0.01708	0.962	0.00071
11.	Modified Henderson and Pabis	a=1.20563, b=-0.11417, c=-0.09146, k=-0.01627, g=469.77978, h=20.17896	0.982	0.00070
12.	Aghabashlo model	k ₁ =-0.074, k ₂ =-0.081	0.984	0.00073
13.	Weibull	a=73.6678, b=1.3160	0.961	0.00051
14.	Midilli <i>et al.</i>	a= 0.9996, b = -2.4234×10 ⁻⁴ , k= 0.00946, n = 1.1460	0.969	0.01103

The above results showed that the value of ‘k’ and ‘a’ increased with increasing T. This can be explained by the effect of T on diffusivity and heat transfer during drying. The increase of temperature accelerates the water migration via diffusion mechanism inside the product and water uptake by the surrounding air.

The time required to reach equilibrium moisture content by the sample varied inversely with time. In other words, drying rate was found to increase with corresponding increase in temperature. This can be attributed to the higher mass transfer rates at elevated temperatures. Moisture ratio was plotted with respect to time at various drying temperatures. It was observed that the moisture ratio decreased with increasing time.

Effective moisture diffusivity of elephant apple

The effective moisture diffusivity was calculated using Fick’s second law of diffusion (Eq.10) by plotting ln(MR) versus time (in minutes) and shown in Figure 2 . The effective diffusivity was evaluated with the method of slopes. The curves are fitted to straight line showed that liquid diffusion is the driving force regulating the drying process. The effective moisture diffusivity values for drying fruit pulp was found to be 1.0947×10⁻¹⁰, 1.173×10⁻¹⁰, 1.348×10⁻¹⁰ and 2.283×10⁻¹⁰ at drying temperatures 50, 60, 70 and 80°C respectively. Moisture diffusivity for the fruit sample exhibited a minimum value of 1.0947×10⁻¹⁰ m²/s at a temperature of 50°C and a maximum value

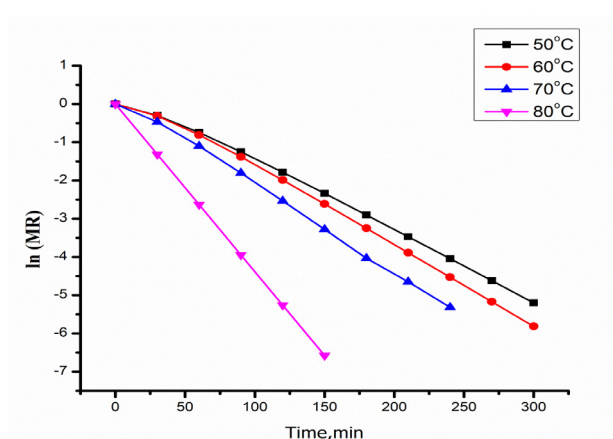


Figure 2. A plot of logarithmic moisture ratio versus drying time for elephant apple at different drying temperatures

of 2.283×10⁻¹⁰ m²/s at 80°C. Comparable values of moisture diffusivity have been mentioned in literature, for instance, 1.57 to 3.96 ×10⁻¹⁰ m²/s for Riesling grape seeds at 40-60°C (Roberts *et al.*,2008), 2.62×10⁻¹⁰ to 4.39×10⁻¹⁰m²/s for raw mango slices at 50-65°C (Goyal *et al.*, 2006),1.264 x 10⁻¹⁰- 4.56 x 10⁻¹⁰m²/sec for jackfruit pulp(Saxena and Dash,2015), 2.231–6.909×10⁻¹⁰ m²/s for white mulberry at 50°C (Doymaz, 2004b) and 1 to 3×10⁻¹¹ m²/s for apricot at 5–80°C (Abdelhaq and Labuza, 1987). It was evident from the results that moisture diffusivity increased with increasing temperatures. Increased temperature results increase in the average energy for transitional, rotational and vibrational motion of vapour resulting in higher moisture gradient and increased mass transfer rate and hence increases

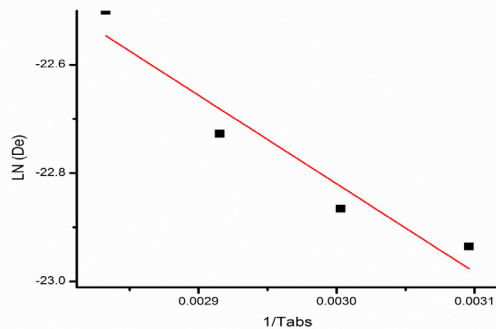


Figure 3. Plot of logarithmic of effective moisture diffusivity ($\ln D_e$) versus reciprocal of absolute temperature ($1/T$), used for calculation of Activation energy.

moisture diffusivity.

Activation energy for drying of elephant apple

The effect of temperature on the diffusivity was expressed through the Arrhenius equation as given by Eq.12. For estimation of activation energy, the logarithms of the calculated effective diffusivities were plotted versus the reciprocal of the absolute temperature and presented in Figure 3. Activation energy of a sample relates to the minimum amount of energy that needs to be provided to trigger moisture diffusion through the sample. Activation energy for the sample was computed as the slope of the linearized plot of logarithmic 'De' versus reciprocal of temperature (Eq. 13 and 14). Activation energy for elephant apple was obtained as 21.95 kJ/mol. Comparable values have been reported in literature for various fruits and vegetables as 28.40 kJ/mol for Green Peas (Simal *et al.*, 1996), 17.40–32.94 kJ/mol for Tomatoes (Doymaz, 2007), 23.02 kJ/mol to 28.10 kJ/mol for carrot slices (Aghbashlo *et al.*, 2011), 22.23 kJ/mol for mushroom (Tulek, 2011), 38.78 kJ/mol for gooseberry (Vega-galvez, 2014), 16.10 kJ/mol for stone apple (Rayaguru, 2012) and 27.22 kJ/mol for kachkal banana peel (Khawas *et al.*, 2014). Pre-exponential factor of Arrhenius equation was calculated as $3.50 \times 10^{-9} \text{ m}^2/\text{s}$. The pre-exponential factor (D_0) in Arrhenius equation represents the diffusivity constant equivalent to the diffusivity at infinitely high temperature.

Conclusion

The drying behavior of elephant apple in a laboratory dryer was investigated at four different drying air temperatures. In order to explain the drying behavior of elephant apple the thin layer drying models were applied and fitted to the experimental data. From the experiment carried out, it can be

satisfactorily stated that the two term exponential model could best explain the thin layer drying behavior of elephant apple pomace. The moisture transfer can be well described by diffusion and the temperature dependence of the effective moisture diffusivities was shown to follow an Arrhenius relationship. Effective moisture diffusivity of the pulped fruit varied from a lowest of 1.095×10^{-10} at 50°C to a highest of $2.283 \times 10^{-10} \text{ m}^2/\text{s}$ at 80°C. Increase in air temperature led to a resultant increase in moisture diffusivity. The rate of drying was found to increase with increasing drying air temperatures. The activation energy, for elephant apple was reported as 21.950 kJ/mol.

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