

EMPIRICAL MODELING OF THIN LAYER DRYING CHARACTERISTICS OF *NAUCLEA LATIFOLIA* LEAVES

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ABSTRACT

The thin layer drying characteristics of *Nauclea latifolia* leaves were studied at four drying temperatures of 35, 45, 55 and 65°C and a constant air velocity of 1.5 m/s in a convective dryer. Experimental kinetic data were fitted to four established drying models available in the literature, namely: the Newton, Henderson and Pabis, Page and Logarithmic models. Model parameters were determined by using non-linear regression analysis while the goodness of fit was assessed by the coefficient of determination (R^2), root mean square error (RMSE) and the standard error (SE). Fick's diffusion model and Arrhenius-type equation were used to determine the effective diffusivity and activation energy, respectively. The increase in air temperature significantly reduced the drying time of the *Nauclea latifolia* leaves. Among the models proposed, the Page model was found satisfactory for describing the air-drying kinetics of *Nauclea latifolia* leaves. The effective diffusivity increases as temperature increases and ranged between 3.3841×10^{-8} - 1.1202×10^{-7} m²/s while the activation energy of diffusion was estimated to be 40.55 kJ/mol.

Keywords: *Nauclea latifolia* leaves, Convective drying, Modeling, Activation energy, Moisture diffusivity.

1. INTRODUCTION

Nauclea latifolia, a member of plant family Rubiaceae, is known to be of high medicinal value (Deeni and Hussain, 1991). *Nauclea latifolia* often known as African peach or pin cushion tree is a straggling evergreen, multi-stemmed shrub or small tree native to tropical Africa and Asia. The leaves, stems, roots and fruits of *Nauclea latifolia* are of high, significant medicinal importance. The extracts of *Nauclea latifolia* have been reported to be potent against bacterial (Okwori *et al.*, 2008; Doughari, 2008), viral (Donalisio *et al.*, 2013), diabetic (Gidado and Ameh, 2005), and plasmodia (Benoit-Vical *et al.*, 1998) activities. The phytochemicals found in *Nauclea latifolia* are indole alkaloids, triterpenes, steroids and saponins (Donalisio *et al.*, 2013). Traditional use of the various parts of the plant include the treatment of jaundice, yellow fever, rheumatism, abdominal pains, loss of appetite, malaria, diarrhea, dysentery, hypertension, diabetes and hepatitis (Donalisio *et al.*, 2013). Ayeleso *et al.* (2014) also reported that *Nauclea latifolia* has strong antioxidant potentials with the leaves demonstrating higher *in vitro* antioxidant activities than the fruits. The leaves were found to contain polyphenols, flavanol, and flavonol which further encourage the use of the leaves of *Nauclea latifolia*. The high moisture content of the leaves

however, makes it perishable and significantly affects its shelf life and quality. To preserve the quality (including bio-activities) of the leaves for effective utilization at any time and enhanced shelf-life, postharvest processing aspect is very important (Rayaguru and Routray, 2011). One of the oldest and most widely employed technologies to avoid spoilage and improve shelf life is drying.

Drying is a method of reduction of water activity values through moisture removal to achieve physicochemical and microbiological stability (Nag and Dash, 2016). Drying removes moisture through simultaneous heat and mass transfer and it provides longer shelf life, lighter weight for transportation and smaller space for storage (Karimi *et al.*, 2012). However, drying is not only affecting the water content of the product, but also alters other physical, biological and chemical properties such as aroma, bioactivity, colour, antioxidant activities as well as palatability of foods (Rayaguru and Routray, 2011; Wojdyło *et al.*, 2009). Therefore, appropriately controlled drying of the leaves appears to be the only means of preserving the quality of the leaves. Hence standardization of drying operation specific to leaves of *Nauclea latifolia* is a necessity in order to achieve the simultaneous purpose of quality preservation and

increased self-life. Literature revealed the documentation of drying behaviour of some medicinal plants such as *Adathoda vasica* leaves (Ganesapillai *et al.*, 2015), ginger and Javanese pepper (Tambunan *et al.*, 2001), rosemary leaves (Arslan and Özcan, 2008), mint leaves (Doymaz, 2006), meadowsweet (*Filipendula ulmaria*) and willow (*Salix alba*) (Harbourne *et al.*, 2009) and bay leaves (Demir *et al.*, 2004). However, investigations on the drying characteristics of *Nauclea latifolia* leaves are scarce in the literature.

The traditional method of drying agricultural products is sun drying. However, the drying time by this method can be quite long, it has associated problems of contamination with dust, soil, sand particles and insects (Afolabi, and Agarry, 2014) and also it is weather dependent and hence difficult to control. Therefore, the drying process should be undertaken in closed equipment to improve the quality of the final product (Ertekin and Yaldiz, 2004; Doymaz, 2006). Hot air drying method has been widely investigated in the literature and identified by some researchers as a more efficient drying method. This is because heat and mass transfer can be controlled during the drying process to achieve desired product quality (Tunde-Akintunde *et al.*, 2014). Factors having a significant effect on drying characteristics of a particular plant material include drying temperature, relative humidity, product size, pre-treatment method (Tunde-Akintunde and Afon, 2010), air velocity and type of drying equipment.

An important aspect of drying technology is the empirical modelling of the drying process. This is because it enables design engineers to choose the best operating conditions for the development of drying equipment according to desired conditions. It can as well be used for improving existing drying systems or even for the control of the drying process. Thin layer drying model remains the best model to describe the food drying process since all commercial flow dryers are designed on thin-layer drying principles (Tahmasebi *et al.*, 2010). Many researchers have investigated the drying kinetics of several medicinal plants to determine the best empirical models for describing thin-layer drying. Such plants include coriander (*Coriandrum sativum*) leaf and stem (Silva *et al.*, 2008), mint leaves (Doymaz, 2006), *Adathoda vasica* leaves (Ganesapillai *et al.*, 2015), and *Pandanus amaryllifolius* leaves (Rayaguru and Routray, 2011). Other agricultural products properties of interest which are necessary for designing and modelling the mass transfer processes of dehydration or moisture adsorption during storage are

effective moisture diffusivity and activation energy. The activation energy indicates the minimum amount of energy required to initiate moisture diffusion from a solid matrix (Nag and Dash, 2016). A Literature survey has revealed that there is a dearth of substantial research on the preservation and drying behavior of *Nauclea latifolia* leaves despite its medicinal importance. Therefore, the objectives of this work are to investigate the thin layer drying characteristics of *Nauclea latifolia* leaves as a function of time at fixed air temperatures, fit the drying data into four thin layer models and determine the effective diffusivities and activation energy of moisture diffusion.

2. MATERIAL AND METHODS

Raw materials

Nauclea latifolia plants freshly harvested from the commercial farm of Landmark University, Omu-Aran, Nigeria were used in this study. It was later identified at the botany unit of the Department of Biological Science of the same institution. When received in the laboratory, the leaves were separated from the stem and root by carefully plucking the leaves. The leaves were later cleaned and washed with water to remove the extra soil. Then, the leaves were sanitized by washing with 25% vinegar solution, wiped with a cloth and cut into 10 cm-long pieces. The thickness of the 20 samples was measured using vernier caliper and the average thickness of the leaves was found as 0.45 mm. The leaves were put into polyethylene bag in a domestic refrigerator operating at 4°C, prior to the drying experiment.

Drying procedure

The drying of the leaves was performed in a hot air convective dryer at temperatures of 35, 45, 55 and 65 °C and drying air velocity of 1.5 m s⁻¹. Prior to placing the sample on the tray, the drying system was run for at least 30 min to obtain steady conditions. Then, the sample was placed on the drying tray in a single thin-layer. Samples of approximately 30 g (\pm 0.5g) were individually identified and used for all experimental runs. The moisture loss was recorded at 10 min intervals by discontinuous weighing of the experimental sample on digital scales, with a \pm 0.01g precision, up to a constant weight that is, the samples reached the equilibrium moisture content with conditions of the drying air. Each experiment was performed twice and averages were reported. The initial moisture content of the leaves was determined by using the oven drying method at 105°C for 5 h (AOAC, 1990). The initial moisture content of the *Nauclea latifolia* was 196.96% dry basis (db).

Empirical modelling of drying curves

Four thin-layer empirical drying models, namely: the Newton (Ganesapillai *et al.*, 2015; Ayensu, 1997), the Henderson and Pabis (Doymaz, 2006; Silva *et al.*, 2008), the Page (Rayaguru and Routray, 2011; Nag and Dash, 2016) and the Logarithmic (Tahmasebi *et al.*, 2010; Aregbesola *et al.*, 2015) models were used to analyse and represent experimental drying data and are shown in Table 1. In the models a, c and n are parameters of the models, k is the drying constant for a particular temperature, (min⁻¹), and t is the drying time

(min). During the drying experiment, the moisture ratio of *Nauclea latifolia* leaves was calculated by using Equation (1).

$$\text{Moisture ratio (MR)} = \frac{M - M_e}{M_0 - M_e} \tag{1}$$

where, MR, *M*, *M*₀, *M*_e are the moisture ratio, moisture content at any time, initial moisture content and equilibrium moisture content, respectively.

Table 1: Thin-layer drying models considered

Model name	Model equation	References
Newton	MR = exp(-kt)	Ganesapillai <i>et al.</i> , 2015; Ayensu, 1997
Henderson and Pabis	MR = a exp(-kt)	Doymaz, 2006; Silva <i>et al.</i> , 2008
Page	MR = exp(-kt ⁿ)	Rayaguru and Routray, 2011; Nag and Dash, 2016
Logarithmic	MR = a exp(-kt) + c	Tahmasebi <i>et al.</i> , 2010; Aregbesola <i>et al.</i> , 2015

Curve fittings for thin-layer kinetic models were performed using the solver function in Microsoft Excel adopting the generalized reduced gradient (GRG2) nonlinear optimization code to determine the model parameters. The best fit line with the minimum sum of square errors (SSE) was used as the sole criterion during curve fitting.

The goodness of fit, R², was calculated using Equation 2:

$$R^2 = 1 - \frac{SSE}{SST} \tag{2}$$

where SST is the total corrected sum of square (Walpole *et al.*, 2002).

SSE measures variation due to error or variation unexplained while SST represents variation in the response values that ideally would be explained by the model. SSE and SST were calculated using Equations 3 and 4 respectively.

$$SSE = \sum_{i=1}^n (MR_{exp,i} - MR_{pre,i})^2 \tag{3}$$

$$SST = \sum_{i=1}^n (MR_{exp,i} - MR_{mean})^2 \tag{4}$$

The best fit model was selected on the basis of high coefficient of determination, R²; low root mean square error, RMSE, as given in Equation 5; and standard error, SE, as given in Equation 6.

$$RMSE = \left[\frac{\sum (MR_{pre,i} - MR_{exp,i})^2}{n} \right]^{1/2} \tag{5}$$

$$SE = \left[\frac{\sum (MR_{pre,i} - MR_{exp,i})^2}{n-1} \right]^{1/2} \tag{6}$$

where, *MR*_{exp,*i*} is the experimental moisture ratio at observation *i*, *MR*_{pre,*i*} is the predicted moisture ratio at this observation, *MR*_{mean} is the overall mean moisture ratio of all observations and n is the number of observations.

3. RESULTS AND DISCUSSION

3.1. Influence of drying temperature

Figure 1 shows thin layer drying curves of *Nauclea latifolia* leaves at the different drying air temperatures considered. Drying air temperature has a significant effect on drying behaviour of *Nauclea latifolia* leaves. At higher temperature, the drying time was shorter due to the fast removal of moisture from the leaves (Revaskar *et al.*, 2014). Similar observations were reported for hot air drying of dika nuts and kernels (Aregbesola, 2015) and hot air drying of elephant apple (Nag and Dash, 2016). The total drying times for *Nauclea latifolia* leaves to reach final moisture content were 80, 120, 180 and 290 min at drying temperatures of 65, 55, 45 and 35°C respectively.

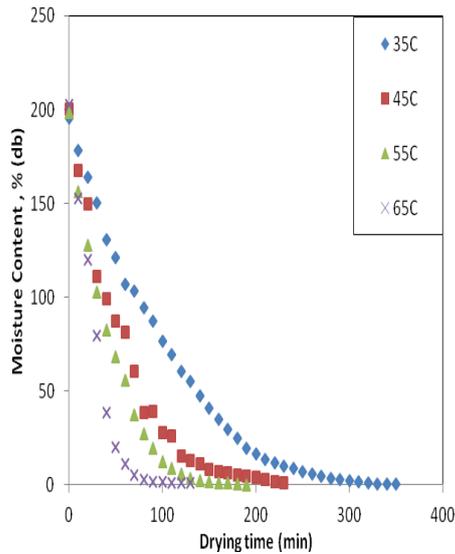


Fig. 1. Moisture content versus time for *Nauclea latifolia* leaves

The graph showing the relationship between moisture ratio and time for *Nauclea latifolia* leaves at all investigated temperature is presented in Fig. 2. It is apparent from the plot that drying occurred predominately during the falling rate period for the leaves. Hence it may be inferred that the dominant physical mechanism governing the moisture movement in *Nauclea latifolia* leaves was diffusion along the moisture concentration gradient as obtained for most agricultural products (Aregbesola *et al.*, 2015).

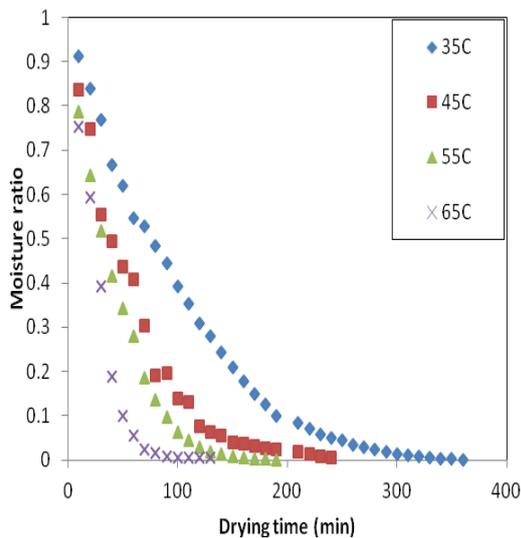


Fig. 2. Moisture ratio versus time for *Nauclea latifolia* leaves

3.2. Evaluation of the models

The empirical drying models of Newton, Henderson and Pabis, Page and Logarithmic were fitted to experimental drying data to determine the model that best fit the data.

The summary of the statistical analysis values used to judge the performance of each model is presented in Table 2. All the tested models described the experimental data well with high coefficient of determination (R^2) values in the range 0.9761 - 0.9960. This is an indication that all the models could satisfactorily describe the air-drying of *Nauclea latifolia* leaves. However, the Page model had the highest R^2 values and the lowest RMSE and SE values compared to other examined thin layer drying models in the range of temperature studied. As indicated in Table 2, the RMSE and SE values of Page model were in the range of 0.017000 - 0.022000 and 0.018000 - 0.023000, respectively. Thus, this model may be assumed to describe best the thin-layer drying behaviour of the *Nauclea latifolia* leaves. Examination of the R^2 , RMSE and SE values showed that Newton model gave the least description of the experimental data with lower R^2 and higher RMSE and SE when compared with other tested thin layer models. The parameters of Page model are as well presented in Table 2.

Both parameter k (drying constant) and n of Page model are in the range of 0.0043825 - 0.0149455 and 1.1061461 - 1.4262697, respectively. Similar ranges of values were reported for sun and solar drying of chilli pepper (Tunde-Akintunde, 2011); hot air-drying of *dika* (*Irvingia gabonensis*) nuts and kernels (Aregbesola, 2015) and vacuum drying of fenugreek, mint, leek and parsley leaves (Zakipour and Hamidi, 2011). Plots of experimental and Page model predicted moisture ratio values with drying time are presented in Fig. 3. As shown in Figure 3, the Page model provided a good description of the experimental kinetic drying data in terms of moisture ratio values. Similar findings were reported by Revaskar *et al.* (2014) for onion slices and Demir *et al.* (2004) for bay leaves.

Table 2: Coefficient of thin layer empirical models and goodness of fit for *Nauclea latifolia* leaves.

Model	Temp (°C)	Parameters	R ²	RMSE	SE
Newton	35	$k = 0.0104$	0.9877	0.032340	0.032799
	45	$k = 0.0182$	0.9913	0.026724	0.027299
	55	$k = 0.0245$	0.9937	0.023131	0.023732
	65	$k = 0.0363$	0.9761	0.049158	0.051014
Henderson and Pabis	35	$k = 0.0108, a = 1.0448$	0.9897	0.029638	0.030058
	45	$k = 0.0186, a = 1.0236$	0.9919	0.025857	0.026413
	55	$k = 0.0238, a = 1.0172$	0.9940	0.022581	0.023167
	65	$k = 0.0379, a = 1.0550$	0.9790	0.046068	0.047807
Page	35	$k = 0.0044, n = 1.1807$	0.9955	0.019622	0.019901
	45	$k = 0.0116, n = 1.1061$	0.9940	0.022000	0.023000
	55	$k = 0.0149, n = 1.1124$	0.9960	0.017000	0.018000
	65	$k = 0.0080, n = 1.4262$	0.9960	0.021000	0.022000
Logarithmic	35	$k = 0.0109, a = 1.0448$ $c = 0.000001$	0.9897	0.029638	0.030059
	45	$k = 0.0186, a = 1.0236$ $c = 0.000001$	0.9919	0.025857	0.026413
	55	$k = 0.0238, a = 1.0172$ $c = 0.000001$	0.9939	0.022581	0.023168
	65	$k = 0.0380, a = 1.0550$ $c = 0.000001$	0.9790	0.046067	0.047807

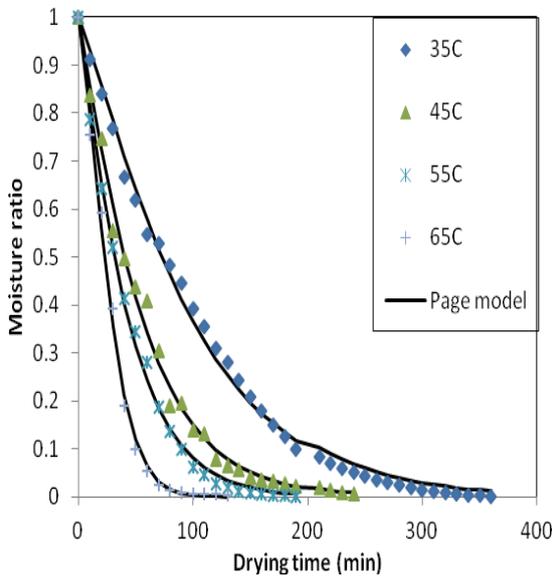


Fig. 3: Influence of air-drying temperature on drying curves and prediction of drying curves by using the proposed Page model.

3.3. Determination of the effective moisture diffusivity

The Fick’s second diffusion equation (Equation 7) was used to interpret the experimental drying data for the

determination of effective diffusivity coefficients at different drying temperatures.

$$\frac{\partial M}{\partial t} = D_{eff} \nabla^2 M \quad (7)$$

Considering the *Nauclea latifolia* plant leaves to conform to slab geometry, the solution of the diffusion equation (Eq.5) assuming: (a) unidimensional moisture movement volume change (b) constant temperature and diffusivity coefficients, and (c) negligible external resistance (Crank, 1975), is presented in Equation 8:

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

For long drying times, according to Doymaz (2006), Equation (8) is simplified to give Equation (9).

$$MR = \frac{8}{\pi^2} \exp \left(- \frac{\pi^2 D_{eff} t}{4L^2} \right) \quad (9)$$

Where D_{eff} is the effective moisture diffusivity (m^2/s), L is the half thickness of the plants leaves of *Nauclea latifolia* (m), and t is drying time (s).

The linearized form of Eq. (9) is expressed in Eq. (10) according to Zakipour and Hamidi, (2011).

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

The effective moisture diffusivity was calculated from the slope (k_o) of the plot of $\ln(MR)$ versus drying time (t) in Figure 4.

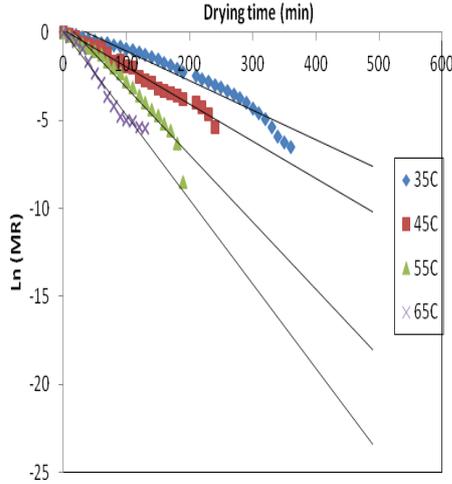


Fig.4. Plot of $\ln(MR)$ versus time for *Nauclea latifolia* leaves.

Table 3: Values of effective diffusivity obtained for *Nauclea latifolia* leaves at different drying temperatures

Temperature (°C)	Effective diffusivity (D_{eff}) (m^2/s)	Equation of fit	R^2
35	3.3841 E -08	- 0.0145x +0.3419	0.9740
45	5.1111 E -08	- 0.0219x +0.1837	0.9812
55	8.0984 E -08	- 0.0347x + 0.4722	0.9812
65	1.1202 E -07	- 0.0480x + 0.1042	0.9681

As indicated in the table, the values of D_{eff} increased greatly as the drying temperature increased. Similar observations were reported for mint leaves (Doymaz, 2006); fenugreek, mint and parsley leaves and vegetative parts of leek leaves (Zakipour and Hamidi 2011); aromatic Pandanus amaryllifolius leaves (Rayaguru and Routray, 2011); elephant apple (Nag and Dash, 2016) and *dika* (*Irvingia gabonensis*) nuts and kernels (Aregbesola *et al.*, 2015). Increase in moisture diffusivity as drying temperature increased has been explained regarding increase in the average energy for transitional, rotational and vibrational motion of vapour which ultimately resulted in higher moisture gradient and increased mass transfer rate and hence increase in moisture diffusivity (Nag and Dash, 2016).

3.4. Activation energy

The temperature dependence of effective diffusivity by Arrhenius relationship is described by Aregbesola *et al.* (2015) as:

$$k_o = \frac{\pi^2 D_{eff}}{4L^2} \quad (11)$$

Table 3 shows the values of D_{eff} obtained for different drying temperatures. Effective diffusivity values ranged from 7.7017 E -09 at 30°C to 1.1202 E -07 m^2/s at 70 °C. These values compare well with values reported for Pandanus amaryllifolius leaves (Rayaguru and Routray, 2011) and mint leaves (Doymaz, 2006). However, the corresponding expressions in Table 3 show that the best fit for each drying temperature is given by a linear relationship ($R^2 > 0.97$).

$$D_{eff} = D_o \exp\left(-\frac{E_a}{RT}\right) \quad (12)$$

where D_o is the pre-exponential factor of the Arrhenius equation (m^2/s); E_a the activation energy (kJ/mol); R the universal gas constant (kJ/mol K) and T is the absolute air temperature (K). The linearized form of Eq. (12) is presented in Eq. (13).

$$\ln(D_{eff}) = \ln D_o - \frac{E_a}{R(T)} \quad (13)$$

Figure 5 shows the plot of $\ln(D_{eff})$ against the inverse of absolute temperature. The graph obtained was essentially a straight line ($R^2 > 0.99$) which indicated Arrhenius relationship. Activation energy of *Nauclea latifolia* leaves (the minimum amount of energy required to initiate moisture diffusion) was calculated from the slope of the straight line described by Arrhenius equation to be 40.55KJ/mol. The corresponding value of the pre-exponential factor of the Arrhenius equation, D_o , which represents the diffusivity constant equivalent to the diffusivity at infinitely high temperature (Nag and

Dash, 2016) was $3.044 \times 10^{-2} \text{ m}^2/\text{s}$. The values of the energy of activation lie within the general range of 12.7 – 110 kJ/mol for food materials (Stamatios and Vassilios 2004).

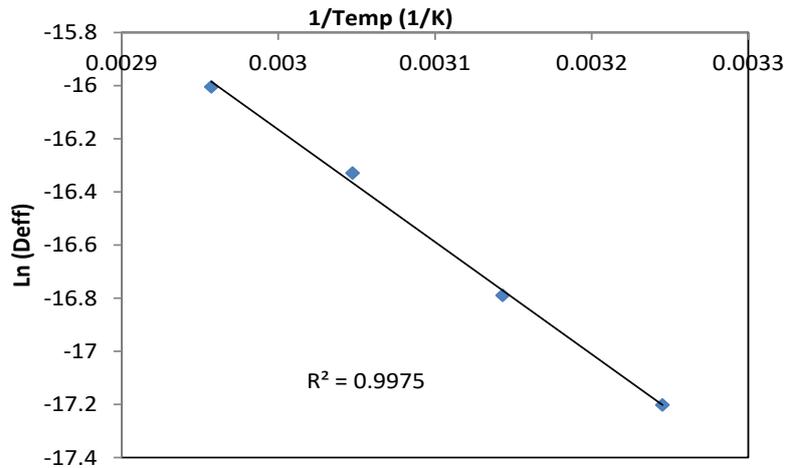


Fig. 5. Influence of air temperature on the effective diffusivity

Table 4 compares the activation energy value obtained for *Nauclea latifolia* leaves with various vegetables and fruits found in literature. It is similar to the activation energy of red pepper drying (Vega-gálvez *et al.*, 2007) and Cape gooseberry (Vega-gálvez *et al.*, 2014), higher than the activation energy of drying elephant apple (Nag and Dash, 2016) and lower than the activation energy of mint leaves drying (Doymaz, 2006) and fever leaves drying (Sobukola and Dairo, 2007).

Table 4: Comparison of activation energy values with literature values

Material	Activation energy (E_a) (KJ/mol)	Reference
<i>Nauclea latifolia</i> leaves	40.55	Present work
Red peppers	40.80	Vega-gálvez <i>et al.</i> (2007)
Mint leaves	62.96	Doymaz (2006)
Fever leaves	80.78	Sobukola and Dairo (2007)
Elephant apple	21.95	Nag and Dash (2016)
Cape gooseberry	38.78	Vega-galvez <i>et al.</i> (2014)

4. CONCLUSION

Thin layer drying characteristics of *Nauclea latifolia* leaves was studied at four drying temperatures in an air

oven. The data obtained was processed and after that fitted into four established semi-empirical equations.

From the study, the following conclusions can be drawn:

- i) The increase in air temperature significantly reduced the drying time of the *Nauclea latifolia* leaves.
- ii) Thin layer drying of *Nauclea latifolia* leaves did not show a constant rate drying period but predominately occurred in the falling rate period; hence, the drying process for *Nauclea latifolia* leaves could be said to be dominantly driven by diffusion.
- iii) The Page model gave the best fit for the description of experimental kinetic data with the highest coefficient of determination (R^2) values and lowest root mean square error (RMSE) and standard error (SE) values compared with other investigated model.
- iv) The values of effective diffusivity for drying at 35 - 65°C of air temperature and 1.5 m/s of air velocity ranged from 3.3841×10^{-8} to $1.1202 \times 10^{-7} \text{ m}^2/\text{s}$. The effective diffusivity increased with the air temperature. The activation energy for moisture diffusion was found as 40.55 kJ/mol.

Nomenclature

a, c	model coefficient
db	dry basis
SE	standard error
RMSE	root mean square error

SSE	sum of square error
SST	total sum of squares
k	drying rate constant (min^{-1})
M	moisture content (% db)
MR	moisture ratio
MR_{mean}	overall mean moisture ratio
M_0	initial moisture content (% db)
M_e	equilibrium moisture content (% db)
$MR_{exp,i}$	i^{th} Experimental moisture ratio
$MR_{pre,i}$	i^{th} Predicted moisture ratio
n	exponent
R^2	coefficient of determination
t	time (min)
D_{eff}	effective diffusivity, m^2/s
D_0	pre-exponential factor of Arrhenius equation, m^2/s
E_a	activation energy, kJ/mol
k_0	slope
R	gas constant
L	half slab thickness

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