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Estimation of effective moisture diffusivity of Red amaranth leaves (*Amaranthus tricolor* L.) for thin-layer drying technology

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Abstract Red amaranth leaves are the excellent source of antioxidant. The chlorinated sample dried at60^0C gave better quality product. Midilli's model was found to satisfactorily describe the drying behavior of red amaranth leaves which showed the highest R^2value, lowest X^2value and RMSE values. The statistical indicators showed that Midilli's model was the best model to describe the drying kinetics of red amaranth leaves.

Keywords: Mathematical modeling, Total phenol, Antioxidant, Moisture diffusivity, Activation energy

Introduction

Amaranths are the most important leafy vegetable of the lowland tropics of Africa and Asia, but are scarcely known in South America (Palada and Crossman, 1999). A tricolor has a variety of leaf colors such as white (light green), dark green, red, purple and variegated (Palada and Chang, 2003) Green and red amaranths are mainly used as leafy vegetables. Red amaranth is a wonderful vegetable with reddish veined dark green leaves or fully red to purple leaves, suitable for growing in warm weather, in which young leaves and stems can be harvested periodically. Red amaranth is also especially nutritious, rich in easily digestible minerals i.e., iron and calcium, as well as protein, vitamin C, and beta-carotene (Islam *et al.*, 2003) The vitamins and minerals present in plants as natural or synthetic antioxidants have been linked to removing harmful molecules called free radicals in the body to help fight against infection and other conditions, including cancer, coronary artery disease, muscular degeneration, and serious eye diseases (Dasgupta and De, 2007). Leafy vegetables are sources of polyphenols, which prevents many chronic diseases, including cancer, cardiovascular diseases, and diabetes, has been well

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documented (Scalbert *et al.*, 2005). Initial findings confirmed that Amaranthus leaves are important sources of antioxidants (Zhou *et al.*, 2012), but it is important to identify the major antioxidants and the antioxidant activity, specifically for red amaranth. Red amaranth cultivars of different flesh color may differ in their phenolic content and antioxidant properties. In addition, cultivars with the same flesh color may differ in total phenolic content and antioxidant activity. Several studies have reported correlations among the antioxidant activities and phytochemical concentrations of various food commodities (Awika *et al.*, 2003); however, this type of information is not available for red amaranth. Information on the total phenolics and antioxidant activity of red amaranth would also be helpful for increasing the awareness of consumers regarding the level of beneficial phyto-chemicals present in this nutritious vegetable.

Leafy vegetables are perishable commodities. Hence optimization of parameters for processing and storage is very important to get best quality product. Maintenance of nutritive value, texture, color and flavour are important in preserving the food. Dehydration is the most inexpensive and oldest preservation techniques of fruits and vegetables. Now a day’s dried foods are very trendy and it’s used as processed or ready- to- eat foods. Dehydrated foods are easy to handle to transport, storage and use.

The objective of this research was the evaluation and the modeling of the drying kinetics of mass transfer during the hot-air drying process of red amaranth leaves, and the analysis of the influence of hot-air dryer conditions on the kinetic constants of the proposed models. The principle of modeling is based on a set of mathematical equations which can satisfactorily explain the system. The solution of these equations must allow calculation of the process parameters as a function of time at any point in the dryer based the primary condition (Kaleta and Górnicki, 2010). Hence, the use of a simulation model is an important tool for prediction of performance of drying systems.

Materials and methods

Preparation of sample for drying

Control sample

Undamaged matured fresh leaves of red amaranth were taken, then properly clean it in tap water for removing foreign matters like mud, dirt, chaff and immature leaves. The surface moisture was removed by using muslin cloth and after that it air dried at room temperature. Then the leaves were dried at three different temperature 50^0C, 60^0C and 80^0C in a tray drier for 4 hour.

Chlorinated sample

Undamaged matured fresh leaves of red amaranth were taken and properly cleaned it in tap water for removing foreign matters like mud, dirt, chaff and immature leaves .The surface moisture was removed by using muslin cloth and then it air dried at room temperature. After that these leaves were dipped in chlorinated water (5 drop of Zeoline-200 in 1L distilled water) for 5min. Then, the leaves were drained out and the surface water on the wet red amaranth leaves and removed with a muslin cloth. The red amaranth leaves were dried at three different temperature 50^0C, 60^0C and 80^0C in a tray drier for 4 hours.

Water blanched sample

Undamaged matured fresh leaves of red amaranth were prepared as mentioned above. These leaves were blanched at 80^0 C temperatures for 3min. After that the blanched leaves were placed under running tap water immediately to cool in ambient temperature and the surface water on the wet leaves of red amaranth was removed with a muslin cloth. The red amaranth leaves were dried at three different temperatures 50^0C, 60^0C and 80^0C in a tray drier for 4hours.

The samples obtained after drying at different temperatures were subjected for further studies.

Analytical parameters

Moisture Content was calculated according to the method described by Ranganna (1986) and expressed as dry basis. Total phenol content was calculated by Folin-ciocaltue method at a wavelength of 765nm using gallic acid standard described by Singleton and Rossi (1965) and expressed as mg of Gallic acid/mg of red amaranth leaves in dry basis. Antioxidant capacity of red amaranth leaves was calculated by FRAP(ferric reducing/ antioxidant power) assay at a wavelength of 593nm using a spectrophotometer by Benzie and strain as modified by pulido (Benzie and strain, 1996) and expressed as mg of ascorbic acid/ 100gm of dry red amaranth leaves. The value of moisture ratio (MR) is calculated by ASAE standard air-oven procedure developed by Henderson and perry (1976) in dry basis.

Drying procedure

A laboratory scale tray dryer was used to conduct this drying experiment. The experiments were conducted in three replications at 50^0C,

60^0C and 80^0C of drying air temperature. Before the experiment, the dryer was allowed to work for 15min to reach steady state at desired temperature. Red amaranth leaves were spreaded uniformly on a tray. The mass loss of the sample was measured by using a digital balance with the accuracy of 0.01g, at15min intervals.

Mathematical modeling

During thin layer drying experiment, the moisture ratio of the red amaranth leaves was determined as follows:-

$MR = \frac{\mathrm{Mt-Me}}{\mathrm{M0-\ Me}}$………………………………. Eq. (A.1)

Where, M_0 is the initial moisture content, M_e is the equivalent moisture content and M_t is the moisture content at time t. All moisture contents are in % of dry basis. From Eq. (A.1) using (Doymaz and Pala *et al.*, 2002) models we can write the expression of moisture ratio as follows:-

$MR = \frac{\mathrm{M_t}}{\mathrm{M_0}}$…………………..…………………… Eq. (A.2)

The different thin layer models and the mathematical expression for these models expressed in Table 1 which used to select the best fitted models for describe the drying curve of red amaranth leaves. Three criteria parameters, R^2 (Coefficient of determination) and RMSE (Root mean square error) and SSE (sum of squares due to error) were used to determine the adequacy of the fit.

Table 1. Six different thin layer drying models applied to describe the drying curve of red amaranth leaves

Model Name	Model Equation	Reference
Newton	$MR = \exp(-kt)$	Liu and Bakker-Arkema (1997)
Page	$MR = \exp(-kt^n)$	Zhang and Litchfield(1991)
Modified page	$MR=\exp(-kt^n)$	Overhults *et al.*(1973)
Henderson and pabis	$MR = a \exp(-kt)$	Henderson and Pebis(1961)
Logarithmic	$MR = a \exp(-kt) + b$	Chandra and Singh (1995)
Midilli	$MR = a \exp(-kt^n) + bt$	Midilli *et al.*(2002)

Moisture diffusivity

Diffusivity explained with Fick's diffusion equation is the only physical mechanism to transfer the water to surface during drying process (Ozbek and

Dadali, 2007; Wang *et al.*, 2007). Effective moisture diffusivity depends on composition, moisture content, temperature and porosity of the material. It was used due to the limited information on the mechanism of moisture movement during drying and complexity of the process (Abe and Afzal, 1998).The coefficient of effective diffusion was determined by using the analytical solution of the equation of the second law of fick for flat slab. The effective moisture diffusivity was calculated by using Crank (1975) equation are as following:-

$$\text{MR} = \frac{Mt-Me}{M_0-M_e} = \frac{8}{\pi^2}\sum_{n=0}^{\propto}\frac{1}{(2n+1)^2}\ \text{exp}\ [-(2n+1)^2\pi^2\ \frac{D_{eff}}{L^2}t] \text{……………..Eq.(A.3)}$$

Where, D_{eff} is the effective moisture diffusivity and expressed as m^2/ s, L is the full thickness of red amaranth leaves and t is the drying time (min), n is a positive integer. For long drying time, the Eq. (3) can be simplified as Eq. (4) by taking the first term of series solution and expressed in alogarithmic form;

$$\text{MR}= \frac{8}{\pi^2}\ \text{exp}\ [-\frac{\pi^2\ .\ D_{eff}}{4L^2}\ \text{t}] \text{ …………………………… Eq.(A.4)}$$

Generally, The moisture diffusivities are calculated by plotting ln(MR) versus drying time to calculate effective diffusivity, a plotted non- linear relationship between the drying time and ln (MR) gives a straight line with a slope of:

$$\text{lnMR} = \ln(\frac{8}{\pi^2}) - (\frac{\pi^2\ .\ D_{eff}}{4L^2}\text{t}) \text{………………………… Eq. (A.5)}$$

$$\text{Slope}=\pi^{2}\frac{D_{eff}}{4l^2} \text{ …………………………… Eq. (A.6)}$$

The empirical values of effective moisture diffusivity (D_{eff}) for different temperature for different pretreated red amaranth samples showed in Table C.

Activation energy

In diffusivity model the activation energy was calculated by the Arrhenius equation (1889)

$$D_{eff}= D_0\ \text{exp}\ (-\frac{E_a}{R.T}) \text{ …………………………………….Eq. (A.7)}$$

Where, D_0 is Arrhenius constant (m^2/s), R is the universal gas constant (8.314*10^-3 KJ/mol. K), Ea is the activation energy and T_{abs} is the absolute temperature (K). The value of activation energy was expressed as the graphical representation of ln (D_{eff}) against 1/T_{abs}. The activation energy was calculated using the Arrhenius equation Eq. (A.6). Logarithm of D_{eff} as a function of the reciprocal of absolute temperature (T) was plotted.

Results

Drying curve

Moisture ratio vs time curves for thin-layer drying techniques at 50^0 C, 60^0 C and 80^0 C temperatures are shown in Figure 1, 2 and 3, respectively. At 50^0 C curves showed the decreasing trend of drying process. After completion of drying maximum moisture ratio was observed with chlorinated sample more non-blanching sample than water blanching sample. Similar result was observed at 60^0 C. At 80^0 C moisture ratios was observed with chlorinated, control more water blanching sample.

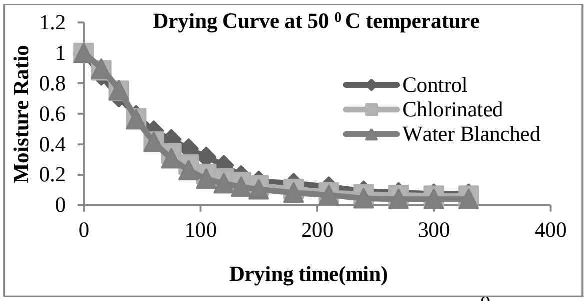

Figure 1. Drying kinetics of red amaranth leaves at 50^0 C temperature

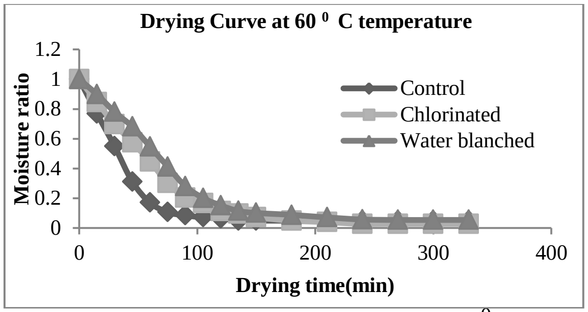

Figure 2. Drying kinetics of red amaranth leaves at 60^0 C temperature

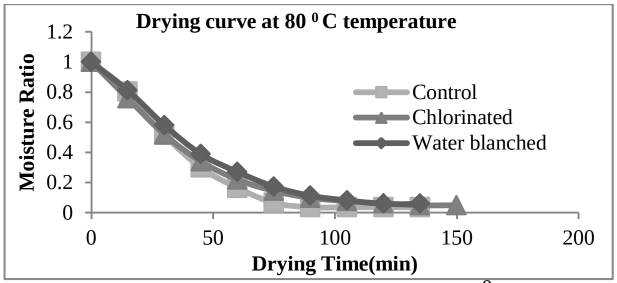

Figure 3. Drying kinetic of red amaranth leaves at 80^0 C temperature

The effect of thermal treatment on total phenol content of Control, chlorinated and water blanched red amaranth leaves sample at 50^0 C, 60^0 C and 80^0 C temperature were shown in Figures 4, 5 and 6 respectively. At 50^0 C temperature control red amaranth leaves sample was the higher total phenol content than chlorinated and water blanching sample after completion of drying. Similar result was observed at 60^0 C and 80^0 C.

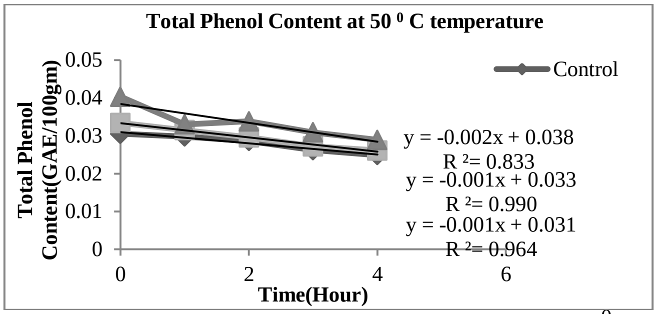

Figure 4. Effect of dehydration of red amaranth leaves at 50^0 C on total phenol content

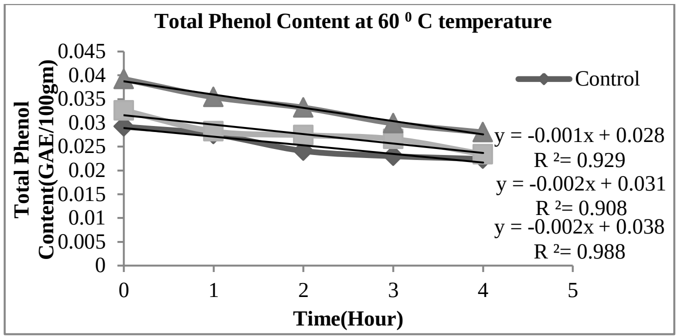

Figure 5. Effect of dehydration of red amaranth leaves at 60^0 C on total phenol content

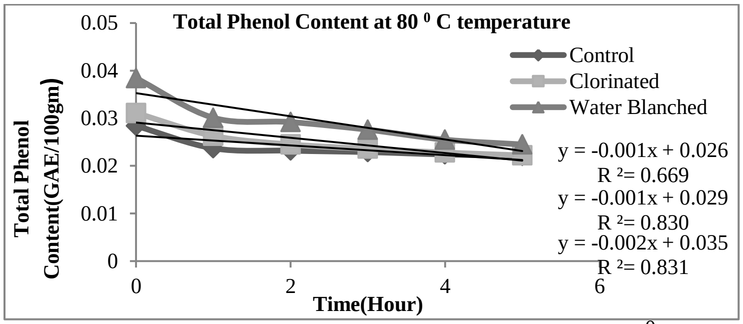

Figure 6. Effect of dehydration of red amaranth leaves at 80^0 C on total phenol content

The effect of thermal treatment on antioxidant capacity of control, chlorinated and water blanched red amaranth leaves sample at 50^0 C, 60^0 C and 80^0 C were shown in Figures 7, 8 and 9. At 50^0 C, control red amaranth leaves sample was the highest antioxidant capacity than chlorinated and then the water blanched sample after completion of drying. Similar result was observed at 60^0 C and 80^0 C.

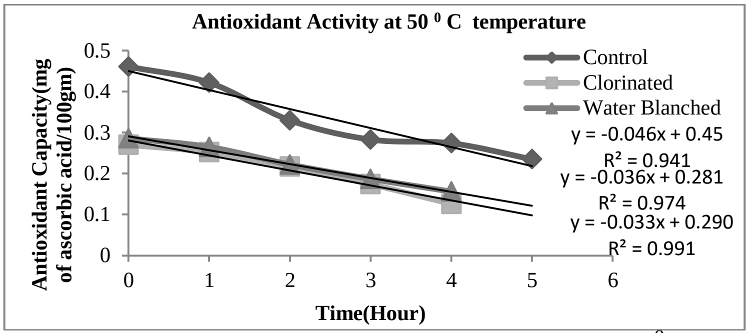

Figure 7. Effect of dehydration of red amaranth leaves at 50^0 C on antioxidant activity

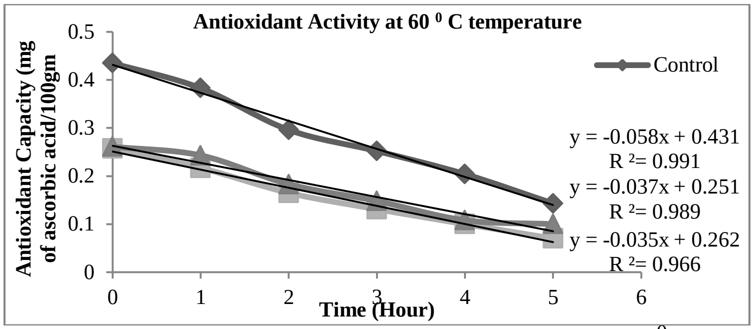

Figure 8. Effect of dehydration of red amaranth leaves at 60^0 C on antioxidant activity

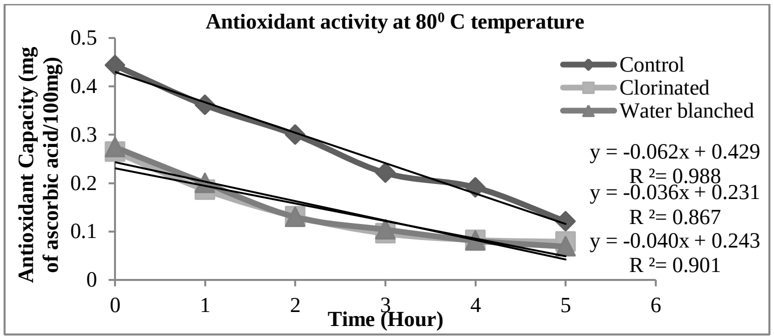

Figure 9. Effect of dehydration of red amaranth leaves at 80^0 C on antioxidant activity

Mathematical modeling for red amaranth leaves

The drying process was completed without further change in weight of the sample. Then moisture content began to decrease from 89.39% to 15%. Later, the moisture content data were converted to moisture ratio and fitted to the six thin layer drying models. The results of statistical analysis for mathematical models of pre-treated red amaranth leaves were shown in Table 2. The best fitted model depended on the highest R^2 value, lowest X^2 value and RMSE values. On the basis of the empirical result for all the temperature in this process of different pre-treated samples, the Midilli's model is the best fitted model.

Table 2. Results for six empirical thin layer drying models of red amaranth leaves

Model Names	**Temperature (^{0} C)**	**Sample Type**	**Model Coefficients and Constants**	**SSE**	**RMSE**
Newton	50	Control	K=0.0108	0.011	0.026
		Chlorinated	K=0.0133	0.024	0.039
		Water blanched	K=0.0143	0.031	0.044
	60	Control	K=0.0245	0.018	0.042
		Chlorinated	K=0.0152	0.022	0.041
		Water blanched	K=0.0124	0.052	0.061
	80	Control	K=0.0262	0.0319	0.059
		Chlorinated	K=0.0235	0.0060	0.024
		Water blanched	K=0.0211	0.0131	0.195
Page	50	Control	K=0.0186, n=0.8797	0.0053	0.02
		Chlorinated	K=0.0088, n=1.0937	0.0221	0.039
		Water blanched	K=0.0048, n=1.250	0.016	0.033
	60	Control	K=0.0091, n=1.259	0.0090	0.033
		Chlorinated	K=0.0046, n=1.2753	0.0045	0.02
		Water blanched	K=0.0019, n=1.425	0.0142	0.033
	80	Control	K=0.0041, n=1.487	0.002	0.02
		Chlorinated	K=0.0143, n=1.127	0.002	0.017
		Water blanched	K=0.0079, n=1.244	0.0017	0.014
Modified page	50	Control	K=0.0108, n=0.879	0.0053	0.02
		Chlorinated	K=0.0133, n=1.0937	0.022	0.039
		Water blanched	K=0.0141, n=1.251	0.016	0.033
	60	Control	K=0.0239, n=1.2586	0.009	0.033
		Chlorinated	K=0.0148, n=1.2753	0.004	0.02
		Water blanched	K=0.0123, n=1.4254	0.014	0.033
	80	Control	K=0.0250, n=1.487	0.0028	0.02
		Chlorinated	K=0.0231, n=1.1267	0.0029	0.017
		Water blanched	K=0.0205, n=1.2441	0.0017	0.014

Table 2. (Con.)

Model Names	Temperature (^{0} C)	Sample Type	Model Coefficients and Constants	SSE	RMSE
Henderson and Pabis	50	Control	K=0.0104, a=0.976	0.0109	0.028
		Chlorinated	K=0.0140, a=1.044	0.0212	0.037
		Water blanched	K=0.0153, a=1.068	0.0231	0.039
	60	Control	K=0.0255, a=1.043	0.0166	0.042
		Chlorinated	K=0.0160, a=1.055	0.0174	0.037
		Water blanched	K=0.0135, a=1.083	0.0392	0.055
	80	Control	K=0.0277, a=1.0652	0.0264	0.057
		Chlorinated	K=0.0241, a=1.0276	0.0050	0.022
		Water blanched	K=0.0220, a=1.0478	0.0099	0.035
Logarithmic	50	Control	K=0.012,a=0.938,b=0.0668	0.0012	0.009
		Chlorinated	K=0.015,a=1.019,b=0.0411	0.0147	0.032
		Water blanched	K=0.016,a=1.058,b=0.015	0.0220	0.039
	60	Control	K=0.025,a=1.046,b=-0.0039	0.0164	0.045
		Chlorinated	K=0.0146,a=1.082,b=0.037	0.014	0.037
		Water blanched	K=0.0126,a=1.1047,b=-0.028	0.0376	0.056
	80	Control	K=0.0243,a=1.106,b=-0.053	0.021	0.056
		Chlorinated	K=0.024,a=1.028,b=-0.0012	0.005	0.024
		Water blanched	K=0.0194,a=1.09,b=-0.055	0.007	0.032
Midilli	50	Control	K=0.020,a=1.065,b=0.0001	0.0007	0.008
		Chlorinated	K=0.012,a=1.121,b=0.0002,n=1.067	0.0033	0.017
		Water blanched	K=0.008,a=1.1003,b=0.0002,n=1.182	0.0037	0.017
	60	Control	K=0.0024,a=0.927,b=0.0004,n=1.619	0.0014	0.015
		Chlorinated	K=0.0015,a=0.927,b=0.00016,n=1.522	0.0019	0.014
		Water blanched	K=0.00038,a=0.9355,b=0.00027,n=1.79	0.0026	0.016
	80	Control	K=0.0028,a=0.99,b=0.00024,n=1.59	0.0002	0.009
		Chlorinated	K=0.011,a=1.0206,b=0.0002,n=1.207	0.000042	0.003
		Water blanched	K=0.0071,a=1.023,b=0.00026,n=1.29	0.00017	0.006

Moisture diffusivity

The moisture diffusivity of red amaranth leaves at three different temperatures for control, chlorinated and water blanched sample was shown in Table 3. The graphical representation of moisture diffusivity at various temperatures for control, chlorinated and water blanched red amaranth sample was shown in Figures 10, 11 and 12.

Table 3. Moisture diffusivity of red amaranth leaves at different drying temperature

Drying Temperature	**Sample**	**Moisture Diffusivity(m^2/s)**
	Control	-3.2456*10^-7
50^0C	Chlorinated	-3.65*10^-7
	Water blanched	-4.057*10^-7
	Control	-8.925*10^-7
60^0C	Chlorinated	-6.491*10^-7
	Water blanched	-4.868*10^-7
	Control	-11.77*10^-7
80^0C	Chlorinated	-8.519*10^-7
	Water blanched	-9.331*10^-7

In our study, three different temperatures of 50^0 C, 60 ^{0} C and 80 ^{0} C were used for tray drying of red amaranth leaves. Control, chlorinated and water blanched samples were used for this drying process. Increased in drying temperature increased the moisture diffusivity of all samples (Table 3).The moisture diffusivity of control sample was 3.2456*10^-7, 8.925*10^-7 and 11.77*10^-7 at 50 ^{0} C, 60 ^{0} C and 80 ^{0} C respectively. The moisture diffusivity of chlorinated sample was 3.65*10^-7, 6.491*10^-7 and 8.519*10^-7 at 50 ^{0} C, 60 ^{0} C and 80 ^{0} C respectively. The moisture diffusivity of processed sample was 4.057*10^-7, 4.868*10^-7 and 9.331*10^-7 at 50 ^{0} C, 60 ^{0} C and 80 ^{0} C respectively. Similar result was reported by Kadam *et al.* (2011) for thin layer convective drying of mint leaves. The moisture diffusivity (D_{eff}) of mint leaves varied from 1.2325×10^{-10} to $2.6568 \times 10^{-10} m^2/s$ for temperature range from 45 to 65^0C. Darvishi *et al.,* (2016) reported the moisture diffusivity of dill leaves as $3.92x10^{-7}$, $4.19x\ 10^{-7}$, $6.22x10^{-7}$ and $7.42x10^{-7}$ for temperature range from 40, 50, 60 and 70 ^{0} C. These values are consistent with the present estimated D_{eff} values for red amaranth leaves. The graphical representation of moisture diffusivity of red amaranth leaves at three different temperatures and three pre treated samples control, chlorinated and water blanched are shown in Figures 10-12.

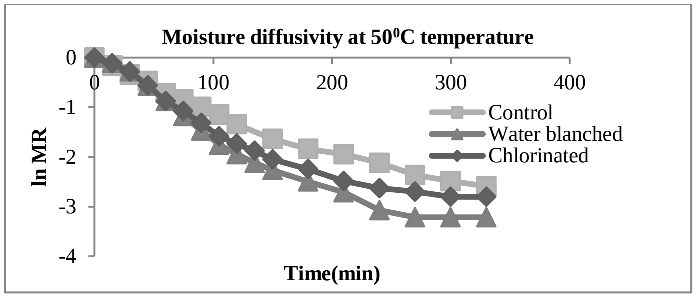

Figure 10. Moisture diffusivity of red amaranth leaves at 50^0 C

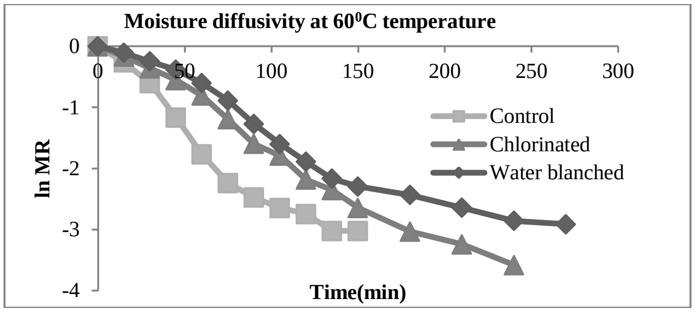

Figure 11. Moisture diffusivity of red amaranth leaves at 60^0 C

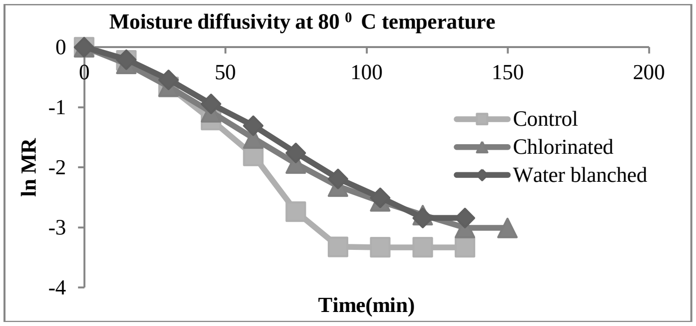

Figure 12. Moisture diffusivity of red amaranth leaves at 80^0C

Activation energy

The activation energy was expressed as the graphical representation of ln (D_{eff}) against $1/T_{abs}$. The activation energy for diffusivity model was determined by the Arrhenius equation (1889):-

$D_{eff} = D_0 \exp(-Ea/RT)$

And slope=$\pi\ ^{2}\frac{D_{eff}}{4l^2}$

From the slope of the straight line described by the Arrhenius equation, the activation energy, *Ea* was determined as 11.94 KJ/mol for Control sample, 12.09KJ/mol for chlorinated sample and 11.43KJ/mol for water blanched sample. Similar result was reported by Kakade and Hathan, (2014) for thin layer convective dehydration kinetics of beet root leaves. The activation energy of blanched and unblanched samples was reported as 15.7913 and 20.9221 kJ/mol, respectively for temperature range from 50, 60, 70 and 80^0 C. The activation energy of various temperatures for chlorinated, water blanched and control samples were shown in Figure 13.

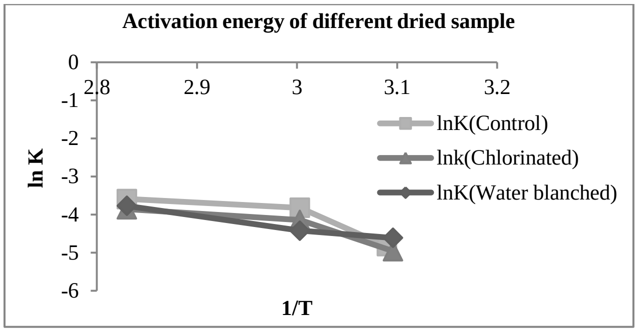

Figure 13. Activation energy of red amaranth leaves dried at different temperatures

Discussion

In this study, six thin-layers drying models namely Newton, Page, Modified page, Henderson and pabis, Logarithmic and Midilli models were used to select a suitable form of the drying curve of red amaranth leaves. The experimental data were used to select the best fitted model to describe the drying kinetics. The best fitted models depended on the basis of lower sum squared errors (SSE) value and root mean square error (RMSE) and the higher value of correlation coefficient.

Values were obtained in Midilli model. The Midilli model described the drying curve of red amaranth leaves satisfactorily. The Midilli *et al.* (2002) model gave the highest value of R^2 (0.9997). For the control, chlorinated sample and water blanched sample at 50^0C showed the lowest SSE (0.0014) and RMSE (0.015), the lowest SSE (0.0019) and RMSE (0.014) for chlorinated sample and the lowest SSE (0.0026) and RMSE (showed 0.016) for water blanched sample, respectively.

At 80^0C temperature the Midilli's model showed the highest value of R^2 (0.99952), the lowest SSE (0.0002) and RMSE (0.009) for control, the highest value of R^2 (0.99952), the lowest SSE (0.000042) and RMSE (0.014) for chlorinated sample and the highest value of R^2 (0.99952), the lowest SSE (0.00017) and RMSE (0.006) for water blanched sample.

Result showed the activation energy of blanched and unblanched samples for temperature range from 50, 60, 70 and 80 ^{0} C was reported as 15.7913 and 20.9221 kJ/mol respectively. Darvishi *et al.* (2016) reported the activation energy of dill leaves was16.84 kJ/mol for temperature range from 40, 50, 60 and 70 ^{0} C. During water blanching water can be removed from the sample by less energy due to the cell rupture of skin. The activation energy of chlorinated sample is higher than the control and water blanched sample. These values are consistent with the present estimated D_{eff} values for red amaranth leaves.

The statistical indicator showed that the Midilli's model was the best fitted model to explain the drying kinetics of red amaranth leaves. Among all these models we can consider the Midilli's model is the best fitted model to explain the drying process of different pre-treated red amaranth leaves sample, because in most cases, it represents the lower RMSE value. The similar results were found in bay leaves (Gunhan *et al.*, 2005) and coriander leaves (Akpinar 2010), Spinach (Doymaz, 2009) and mint leaves (Doymaz, 2006). Akpinar (2005) presented similar findings to the ones reported above regarding apples and pumpkin. (Venkanna *et al.*, 2019) also represents the similar result for coriander (Coriandrum sativum) leaf.

When the drying temperature is raised in thin layer drying process for treated and untreated red amaranth leaves, the drying time is diminished and the drying rate is enhanced. The Midilli's model is the best empirical mathematical model to represents the drying curve of thin layer drying process of treated and untreated red amaranth leaves sample for all the drying temperature. In thin layer drying process blanching applied as a pretreatment to reduce the drying rate of the leaves.

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