

Thin Layer Drying Kinetics of of Roselle

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Abstract: This study was performed to determine the most appropriate thin layer drying model and the effective moisture diffusivity of Roselle (*Hibiscus sabdariffa*). Roselle with an Initial Moisture Content (IMC) of 85%, on wet basis (wb) was dried in a conventional tray dryer at temperatures of 40, 50 and 60°C. The drying data were fitted to eleven thin layer models and a thin layer model for the roselle calyx was developed by regressing the coefficients of the best fit model. The newton model was most adequate model for describing the thin layer drying kinetics of the roselle calyx. The drying constant was found to vary linearly with temperature. Also, effective diffusivity was evaluated by using Fick's second law, which varied from 1.405×10^{-10} to 2.283×10^{-10} m²/s. The dependence of moisture diffusivity on temperature was described by Arrhenius type equation. The diffusivity constant D_0 activation energy E_a could be, respectively, estimated as 4.5×10^{-7} m²/s and 21.02 kJ/gmol.

Key words: Activation energy, diffusivity, drying, moisture ratio, roselle, thin layer drying

INTRODUCTION

Roselle (*Hibiscus sabdariffa*) is a tropical shrub found around the world with an approximate height of three meters. Roselle calyx is utilized in the processing of fruit preserves, jellies and jams for its rich content in pectin, ascorbic acid and anthocyanin color (Mouning and Badrie, 2006; Pouget *et al.*, 1990; D'Heureux and Badrie, 2004). In addition, due to their anthocyanins content, pectins, and compounds responsible for the flavor and aroma (Hall, 1968; Ray, 1988; Beristain *et al.*, 1994; Esselen and Sammy, 1975) as well as their antioxidant, diuretic, digestive, and sedative properties (Akindahunsi and Olaleye, 2003; Carbajal *et al.*, 2005).

Roselle calyx is usually harvested at high moisture content (85%, wet basis). Therefore, drying is an important post-harvest treatment prior to reduce the moisture content and to increase the shelf life. Drying is a process comprising simultaneous heat and mass transfer. Many mathematical models have been used to describe the drying process. A considerable amount of work has been done on thin layer drying of different agricultural products. Some of the thin layer models reported were for drying of rapeseed (Duc *et al.*, 2011), litchi (Janjaia *et al.*, 2011), sorghum (Shen *et al.*, 2011), and finger millet (Radhika *et al.*, 2011).

Actually, the thin layer drying model was applied in drying of rosella (Saeed *et al.*, 2008). However, this study did not determine the effective moisture diffusivity and the temperature dependence of diffusivity. Therefore, the objective of this study was to determine the thin layer drying kinetics of roselle calyx, namely the thin layer

drying model, effective moisture diffusivity, and activation energy of roselle calyx. A simple diffusion model based on Fick's second law of diffusion was considered for the evaluation of effective moisture diffusivity. An Arrhenius type equation was applied to determine the temperature dependence of diffusivity.

MATERIALS AND METHODS

The experimental studies were carried out in laboratory of Department of Chemical Engineering, University of Diponegoro, Semarang, Indonesia.

Drying experiments: Fresh calyces of Roselle (with 85% moisture content, w.b.) were collected from the a field in Semarang. The seed's capsules were removed before commencing the drying experiments, and the calyces were used as whole. The initial and final moisture content of the samples was determined by oven method at 105°C (Ruiz, 2005).

A laboratory tray dryer was used for drying, which consist of a blower, heater, and temperature controller. A convective oven was used to determine the initial and final moisture content. The dimension of the drying chamber were 0.5×0.4×0.4 m.

Roselle samples of 250g were dried in dryer at temperatures 40, 50 and 60°C. The air velocity was 1.5-1.6 m/s, which was measured using anemometer. Moisture loss was measured using digital balance and recorded each 5 min with and accuracy of ±0.01g. When the weight of samples reached almost constant, the experiment was stopped.

Mathematical modeling: The experimental roselle calyx drying data at three different temperatures were fitted using eleven thin layer drying models listed in Table 1. In these models, MR represents the dimensionless moisture ratio namely $MR = (M - M_e) / (M_o - M_e)$, where M is the moisture content at any time, M_o is the initial moisture content and M_e is the equilibrium moisture content. The values of M_e may be relatively small compared to M and M_o , so the equation can be simplified to $MR = M / M_o$ (Radhika *et al.*, 2011).

The non linear regression analysis in the present study was performed using the software MATLAB 7.0. Statistical parameters such as the correlation coefficient (R^2), the chi square (χ^2) and the root mean square error (RMSE) were used to assess the goodness of the fitting. The best fit was that which results in higher R^2 and the lowest χ^2 and RMSE (Duc *et al.*, 2011; Janjaia *et al.*, 2011; Radhika *et al.*, 2011; Shen *et al.*, 2011). The reduced c^2 and RMSE were evaluated as:

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

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

where, $MR_{exp,i}$ is the i^{th} experimentally observed moisture ratio, $MR_{pre,i}$ is the i^{th} predicted moisture ratio, N is the number of observations and Z, the number of constants in models.

Effective diffusivity and activation energy: It has been accepted that the drying characteristics in the falling rate period could be described by using Fick's diffusion equation (Crank, 1975). The form Eq. (3) could be used for particles with slab geometry by assuming uniform initial moisture distribution and for long drying time,

$$MR = \frac{8}{\pi^2} \exp\left[-\pi^2 \frac{D_{eff}}{4L^2} t\right] \quad (3)$$

where D_{eff} is the effective diffusivity (m^2/s); L is the half thick-ness of slab (m). Then, Eq. (3) is written in a logarithmic form as follows:

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

Diffusivity could be typically determined by plotting experimental drying data in terms of $\ln MR$ versus drying time in Eq. (4), and calculated by these slope of the

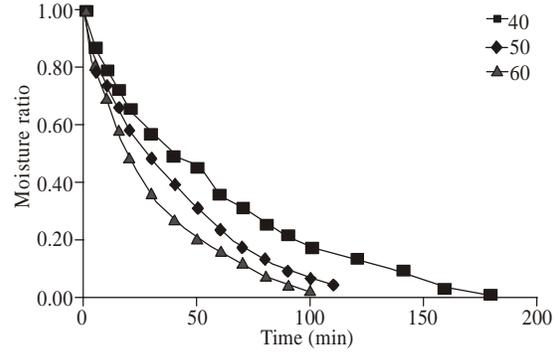


Fig. 1: Drying curve of roselle calyx at different temperatures

providing a straight line. The effective moisture diffusivity could be related with temperature by simple Arrhenius equation as given below (Shen *et al.*, 2011):

$$D_{eff} = D_0 \exp\left(-\frac{E_a}{RT}\right) \quad (5)$$

where D_{eff} is the effective moisture diffusivity (m^2/s), D_0 is the constant equivalent to the diffusivity at infinitely high temperature (m^2/s), E_a is the activation energy (kJ/mol), R is the universal gas constant (8.314 J/(mol K)), and T is the absolute temperature (K). The activation energy (E_a) and the constant (D_0) could be determined by plotting $\ln(D_{eff})$ versus $1/T$ after linearization for Eq. (5).

RESULTS AND DISCUSSION

The changes in moisture content with time for three different drying air temperatures are shown in Fig. 1, which indicated that the moisture ratio decreased with the increased drying time. Further it can be observed that the drying air temperature has an important effect on the drying rate and the total drying process was found to be occurred in the falling rate period only. Therefore, the drying behavior of the Rosella was diffusion governed.

Eleven thin-layer drying models were fitted to the experimental data of moisture ratio of roselle calyx dried at three different temperatures and the fitted thin layer models given in Table 1. The parameter values of R^2 , χ^2 , RMSE and the drying model coefficients were listed in Table 2. It is assumed that the model which has highest R^2 and the lowest χ^2 and RMSE could be considered as the best fit. The Newton model was found to the best, followed by Henderson and Pabis, Two term model, and Two term exponential.

Furthermore, to take into account the effects of drying temperature on the model parameter k in Newton model, and attempting to generalize the model, a regression analysis was applied to set up the relationship between k parameter and the temperature. Then the

Table 1: Thin layer drying models

No	Model name	Equation	Reference
1	Newton	$MR = \exp(-kt)$	Ayensu (1997)
2	Page	$MR = \exp(-kt^n)$	Menges and Ertekin (2006)
3	Modified Page	$MR = \exp(-kt)^n$	White <i>et al.</i> (1981)
4	Modified Page II	$MR = \exp(-c(t/L)^n)$	Midilli <i>et al.</i> (2002)
5	Henderson and Pabis	$MR = a \exp(-kt)$	Kashaninejad <i>et al.</i> (2007)
6	Modified Henderson and Pabis	$MR = a \exp(-kt) + b \exp(-gt) + c \exp(-ht)$	Yaldiz and Ertekin (2001)
7	Logarithmic	$MR = a \exp(-kt) + c$	Togrul and Pehlivan (2002)
8	Two term model	$MR = a \exp(-k_1t) + b \exp(-k_2t)$	Wang <i>et al.</i> (2007)
9	Two-term exponential	$MR = a \exp(-kt) + (1-a)\exp(-kat)$	Sacilik <i>et al.</i> (2006)
10	Simplified Fick's diffusion	$MR = a \exp(-c(t/L^2))$	Diamante and Munro (1991)
11	Diffusion approach	$MR = a \exp(-kt) + (1-a) \exp(-kbt)$	Togrul and Pehlivan (2002)

Table 2: Statistical results of different thin layer drying models

Model	T (°C)	Model constants			R ²	χ ²	RMSE
Newton	40	k = 0.0182			0.9801	0.0009	0.0294
	50	k = 0.0259			0.9882	0.0011	0.0320
	60	k = 0.0335			0.9963	0.0004	0.0187
Page	40	k = 0.3199	n = 0.2792		0.3842	0.0302	0.1632
	50	k = 0.3838	n = 0.2660		0.7003	0.0304	0.1613
	60	k = 0.4392	n = 0.2681		0.7195	0.0309	0.1616
Modified Page	40	k = 0.6119	n = -0.0639		-0.9196	0.0940	0.2881
	50	k = 0.5875	n = 0.0027		0.3132	0.0696	0.2442
	60	k = 0.6255	n = -0.0339		0.2224	0.0855	0.2690
Modified Page II	40	c = 0.3199	n = 0.2792		-3.0379	0.1978	0.4178
	50	c = 0.3838	n = 0.2660		-0.8587	0.1882	0.4017
	60	c = 0.4392	n = 0.2681		-1.0627	0.2269	0.4381
Henderson and Pabis	40	a = 0.9118	k = 0.0154		0.9802	0.0010	0.0293
	50	a = 0.9149	k = 0.0223		0.9901	0.0010	0.0293
	60	a = 0.8947	k = 0.0279		0.9867	0.0015	0.0351
Modified Henderson and Pabis	40	a = 0.8131	b = 0.3747	c = 0.5229	-1.5267	0.0967	0.2502
		k = 0.1995	g = 0.0028	h = 0.5875			
	50	a = 0.8019	b = 0.3710	c = 0.5250	-1.0637	0.1058	0.2459
		k = 0.2155	g = 0.0060	h = 0.5849			
60	a = 0.8053	b = 0.3524	c = 0.5376	-1.0506	0.1098	0.2431	
	k = 0.2413	g = 0.0096	h = 0.5786				
Logarithmic	40	a = 0.6471	k = 0.0214	c = 0.1737	0.7830	0.0114	0.0968
	50	a = 0.6473	k = 0.0310	c = 0.1721	0.8951	0.0116	0.0954
	60	a = 0.6384	k = 0.0433	c = 0.1710	0.8831	0.0141	0.1043
Two term model	40	a = 0.8229	k1 = 0.0133		0.9582	0.0024	0.0425
		b = 0.1209	k2 = 0.2965				
	50	a = 0.8205	k1 = 0.0192		0.9825	0.0021	0.0390
		b = 0.1220	k2 = 0.2942				
60	a = 0.8039	k1 = 0.0246		0.9812	0.0025	0.0418	
	b = 0.1228	k2 = 0.2891					
Two-term exponential	40	a = 0.5738	k = 0.0233		0.9673	0.0009	0.0285
	50	a = 0.5705	k = 0.0334		0.9662	0.0012	0.0315
	60	a = 0.5733	k = 0.0432		0.9923	0.0003	0.0149
Simplified Fick's diffusion	40	a = 0.9118	c = 0.0154		0.9654	0.0010	0.0293
	50	a = 0.9149	c = 0.0223		0.9706	0.0011	0.0293
	60	a = 0.8947	c = 0.0279		0.6227	0.0141	0.1043
Diffusion approach	40	a = 0.6456	b = 0.2312	k = 0.0321	0.9285	0.0022	0.0421
	50	a = 0.6464	b = 0.2323	k = 0.0460	0.9302	0.0026	0.0452
	60	a = 0.6468	b = 0.2263	k = 0.0618	0.9578	0.0016	0.0349

equations relating the constants of the Newton model with the drying temperature are the following:

$$MR = \exp(-kt), \text{ where } k \text{ is constant}$$

$$k = 0.000765T - 0.012383, R^2 = 0.999986$$

Thus, the thin layer model for Rosella calyx was:

$$MR = \exp[-(0.000765T - 0.012383)t]$$

Figure 2 shows the comparison between the predicted and experimental data of thin-layer drying of roselle calyx at three temperatures for the Newton model. It may be observed from the figure that the agreement between experimental values and predicted values of this model is excellent.

The effective diffusivities of the rosella calyx at three different temperatures was evaluated by plotting $\ln MR$ vs t (Fig. 3) and the data was presented in Table 3. The

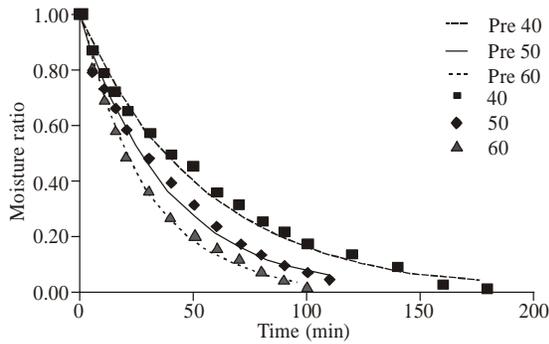


Fig. 2: Drying curves for the experimental data and that predicted based on the Newton model

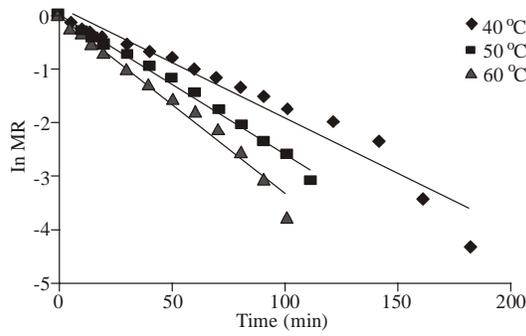


Fig. 3: lnMR vs drying time (min)

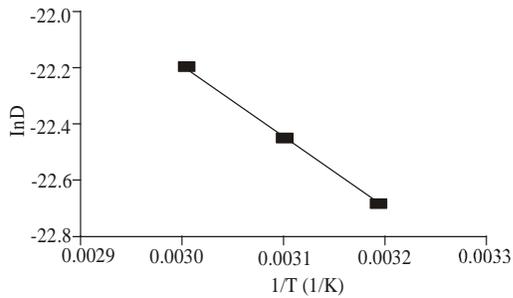


Fig. 4: Variation of effective diffusivity as function of temperature

No	Temperature (°C)	Diffusivity (m ² /s)
1	40	1.405x10 ⁻¹⁰
2	50	1.770x10 ⁻¹⁰
3	60	2.283x10 ⁻¹⁰

values varied from 1.405×10^{-10} to 2.283×10^{-10} m²/s, and it could be obviously found the D_{eff} increased as the temperature increased.

Furthermore, the logarithm of D_{eff} as a function of the reciprocal of absolute temperature was plotted (Fig. 4.). The results showed a linear relationship between (ln D_{eff}) and (1/T), leading to an Arrhenius type relationship between the diffusion coefficient and temperature. According to Fig. 4, the R² for the regression was 0.9979.

Thereby, the diffusivity constant D_0 could be calculated as 4.5×10^{-7} m²/s and the activation energy was evaluated as 21.02 kJ/gmol.

CONCLUSION

The Newton model was the best one to describe drying process of the rosella calyx. The effective diffusivities increased with the drying temperature and varied from 1.405×10^{-10} to 2.283×10^{-10} m²/s. The temperature dependence of diffusivity follows Arrhenius type of relationship. The diffusivity constant D_0 activation energy E_a could be, respectively, estimated as 4.5×10^{-7} m²/s and 21.02 kJ/gmol.

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