# Mathematical modelling of roselle seeds (*Hibiscus sabdariffa* L.) drying kinetics

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# Abstract

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Roselle seeds (Hisbiscus sabdariffa L.) are among the seeds that are high in nutrients and can be useful in several applications. Through the drying process, the seeds can be preserved for a long time. The roselle seeds were oven-dried at different temperatures 55, 60, 65 and 70°C, giving different outlet values for yield and final moisture. A total of five empirical mathematical models (Newton, Henderson and Pabis, Logarithmic, Diffusion approach and Page) were selected to describe and compare the drying characteristics of roselle seeds at respective drying temperatures. The adequately suitable drying temperature was found to be 70°C with a drying time of 4 hrs. The effective moisture diffusivity was calculated using the Fick diffusion equation. The results showed that the moisture content of the seeds gradually decreased with time. The effective diffusivity coefficient of moisture transfer varied from  $5.4262 \times 10^{-10}$  to  $1.074 \times 10^{-9}$  m<sup>2</sup>/s over the temperature range investigated. Mathematical models were fitted to the experimental data and by statistical comparison, the Page model represented drying characteristics better than the other equations with the highest R<sup>2</sup> (0.998) and the lowest values of  $\chi^2$  (0.00033) and RMSE (0.00023) observed for drying air temperature of 70°C. The dependence of moisture diffusivity on temperature was described by the Arrhenius equation, with the estimated activation energy being 43.08 kJ/mol within 55 to 70°C. Based on the selected model, it is possible to predict the moisture change during the drying of roselle seeds and thereby better control the process.

# 1. Introduction

The Roselle plant (Hibiscus sabdariffa L.) is a herbal shrub plant that originated in North Africa and Southeast Asia including Vietnam. Nowadays, they grow in many tropical and subtropical climates. People use various parts of the plant as food and medicine. Apart from their delightful taste, roselle is a good source of nutrients, vitamins and minerals (Jabeur et al., 2017). This plant is grown for the calyces or petals of the flower which are mainly used to prepare herbal drinks, beverages, jam and jellies. After removing the calyces, the capsules containing the seeds are disposed of as a by-product. Usually, the seeds are often discarded, not yet exploited and used effectively. Roselle seeds are considered a potential new source of healthy edible oil (Nyam et al., 2009; Al-Okbi et al., 2017), rich in protein content and micronutrients (Omabuwajo et al., 2000). The chemical and nutritional values of roselle seeds were studied by Samy (1980). Hainida et al. (2008) studied about nutritional and amino acid contents of differently treated

roselle seeds and discovered seventeen essential and non -essential amino acids, they also indicated that roselle seeds can serve as a potential source of functional ingredients.

Human health is built on the foundation of eating habits. General knowledge of the relationship between dietary fibre and health is also an essential issue in today's developed society, so the average intake and the recommended daily intake need to be considered. Roselle seeds not only contain valuable bioactive compounds beneficial to health but also contain dietary fibre. A high amount of crude fibre implied that the roselle seed could be considered a good source of dietary fibre (Omabuwajo et al., 2000; Hainida et al., 2008). Roselle seed might be useful as a low-cost source of dietary fibre substitute in a dietary supplement or food ingredient in the food industry. However, after harvesting, the seeds are also very susceptible to mould in tropical conditions, so drying is also a way to preserve and use the seeds for a long time while maintaining the FULL PAPER

nutrients at the highest concentration. Thin-layer drying is a widely used and suitable technology for by-products. Drying is a complex thermal process in which unstable heat and moisture transfer occur simultaneously (Sahin and Dincer, 2005). This process not only affects the water content of the product but also alters other quality and palatability of the food (Harish et al., 2014). Besides, drying also reduces the cost of packaging, storage and transportation (Vega-Gálvez et al., 2010). Modelling the drying process gives mathematical as well as physical insight into the process. The principle of modelling is based on having a set of mathematical equations that can fully characterize the system. The best drying model should be simple, accurate and applicable in the drying process and need only a short calculation time, which is favourable for quick decision-making in the industry (Thuy et al., 2021). However, limited research has been carried out on the exploitation and utilization of seeds as a potential alternative for human food sources, especially in Vietnam. The detailed research on drying roselle seeds has also not been thoroughly studied. Thus, this research aimed to formulate and validate mathematical а model representative of roselle seeds by thin-layer drying in an oven drier and investigate the goodness of fit. The effective moisture diffusivity and activation energy of roselle seeds during drying was determined.

### 2. Materials and methods

### 2.1 Materials preparation

Roselle was purchased from Dong Thap province (Vietnam). After separating the calyx, the roselle seeds were freshly obtained. Each experiment used approximately 100 g seed and run three replicates.

#### 2.2 Drying experiments

Thin-layer drying experiments with roselle seeds were carried out in an oven drying system (SIBATA, SD-60, Japan) designed for drying agricultural products. The drying process was investigated with temperatures ranging from 55°C to 70°C. The recording of the seed's weight was obtained every 30 mins intervals for up to several hrs until the moisture content was almost constant. Moisture content (MC) on a dry basis (db, %) of dried roselle seeds was calculated as Equation 1 (Ceylan *et al.*, 2007).

$$\% MC_{db} = (W_w/W_d) \times 100 \tag{1}$$

Where  $W_w$  and  $W_d$  are the weight of water and dry matter of roselle seeds (g).

#### 2.3 Mathematical modelling

The drying curves obtained from the experiments were fitted into five mathematical models that are typically used to describe the drying behaviour of thin layers in our study, as shown in Table 1. Data from the drying process of roselle seeds were statistically analyzed using Stagraphics Centurion XVI software to choose the best model.

The moisture content data at various temperatures were transformed to Moisture Ratio (MR), which represents the dimensionless moisture ratio, in order to be suitable with mathematical models (Equation 2).

$$MR = \frac{M_t - M_e}{M_o - M_e}$$
(2)

where M is the instantaneous moisture content (kg water kg<sup>-1</sup> dry matter) of the product,  $M_o$  is the initial moisture content of the product and  $M_e$  is the equilibrium moisture content. The values of  $M_e$  are relatively negligible compared with M and  $M_o$  for a long drying time. Thus, Equation 2 has been simplified to Equation 3 (Toğrul and Pehlivan, 2004).

$$MR = \frac{M_t}{M_o}$$
(3)

Critical statistical calculations such as the correlation coefficient (R<sup>2</sup>), chi-square ( $\chi^2$ ) and root mean square error (RMSE) were used in regression analysis to determine the most suited model for drying thin layer roselle seeds at various temperatures. The best match between experimental data and mathematical models was represented by the highest values of R<sup>2</sup> and the lowest values of  $\chi^2$  and RMSE (Akpinar *et al.*, 2003). Equations 4, 5, and 6 were used to calculate these statistical values (Akpinar, 2010).

Table 1. Mathematical models used to predict the drying of agricultural products.

No.	Models	Equation	References
1	Newton	MR = exp(-kt)	Sobukola <i>et al.</i> (2007)
2	Page	$MR = \exp(-k(t^n))$	Akpinar and Bicer (2008)
3	Henderson and Pabis	MR = a.exp(-kt)	Rosa et al. (2015)
4	Logarithmic	$MR = a.exp(-kt)+b.exp(-k_ot)$	Sobukola et al. (2007)
5	Diffusion Approach	MR = a.exp(-kt)+(1-a)exp(-kbt)	Sobukola et al. (2007)

Where t is drying time (hrs) and a, b, c, h, n, k, k<sub>o</sub> are the model constants.

$$R^{2} = \frac{\sum_{i=1}^{N} (MR_{i} - MR_{pre,i}) \cdot \sum_{i=1}^{N} (MR_{i} - MR_{exp,i})}{\sqrt{\left[\sum_{i=1}^{N} (MR_{i} - MR_{pre,i})^{2}\right] \cdot \left[\sum_{i=1}^{N} (MR_{i} - MR_{exp,i})^{2}\right]}} \quad (4)$$

$$\chi^{2} = \frac{\sum_{i=1}^{N} (MR_{\exp,i} - MR_{pre,i})}{N - z}$$
(5)

$$RMSE = \left[\frac{1}{N}\sum_{i=1}^{N} (MR_{\text{pre},i} - MR_{\text{exp},i})^2\right]^{1/2}$$
(6)

Where  $MR_{exp,i}$  is the i<sup>th</sup> experimentally observed moisture ratio,  $MR_{pre,i}$  is the i<sup>th</sup> predicted moisture ratio, N is the number of observations and z is the number of constants in the model.

# 2.4 Calculation of the effective moisture diffusivity and activation energy

Fick's diffusion equation is a useful way to describe the drying characteristics of diverse foods (John *et al.*, 2014). A variant of the equation given by Crank (1979) can be constructed in the situation of thin-layer drying, assuming one-dimensional moisture transport, minor shrinkage, constant diffusivity, uniform initial moisture distribution, and negligible external resistance (Thorat *et al.*, 2012) (Equation 7).

$$MR = \frac{8}{\pi^2} \sum_{n=0}^{\infty} \frac{1}{(2n+1)^2} \exp\left(-(2n+1)\pi^2 \frac{D_{eff}t}{4L^2}\right)$$
(7)

Where  $D_{eff}$  is the effective diffusivity (m<sup>2</sup>/s), t is drying time (s), n is a positive integer and L is the haft thickness of the slab (m).

For lengthy drying times, the linear solution is derived by assuming that only the first term in the series equation is relevant (n = 0). The natural logarithm of both sides is then used to derive Equation 8 (Akgun and Doymaz, 2005).

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

Because the plot shows a straight line with a slope as Equation 9, the diffusion coefficients can be derived by graphing experimental drying data in terms of ln (MR) vs drying time (t) (Zarein *et al.*, 2015).

$$Slope = \frac{\pi^2 D_{eff}}{4L^2}$$
(9)

The activation energy is computed using the Arrhenius equation (Equation 10), which expresses the influence of drying air temperature on the effective diffusion coefficient (Sanjuán *et al.*, 2003).

$$D_{eff} = D_o exp\left(-\frac{E_a}{RT}\right) \tag{10}$$

where  $D_o$  is the diffusion coefficient corresponding to infinite temperature (m<sup>2</sup>/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 drying air temperature (°K).

### 3. Results and discussion

### 3.1 Drying kinetics of roselle seeds

The effect of temperature on thin-layer drying of roselle seeds was most noticeable with the moisture ratio decreasing continuously with increased temperature and time (Figure 1). It could be observed that the drying rates were highest at the beginning of drying when the moisture content was the greatest and thereafter drying speed is reduced. The drying rate was higher for higher air temperatures, as a result, the time taken to reach the final moisture content was less, the final moisture content of roselle seeds reached 1.57 to 1.80% (DW) after 6.5, 6, 5 and 4 hrs, respectively at drying temperatures of 55, 60, 65 and 70°C. Similar results were obtained by several investigators, they reported a significant increase in the drying rates when higher temperatures were used for drying various agricultural products such as broccoli (Doymaz, 2013), canola (Gazor and Mohsenimanesh, 2010) and Butterfly pea flowers (Thuy et al., 2021).

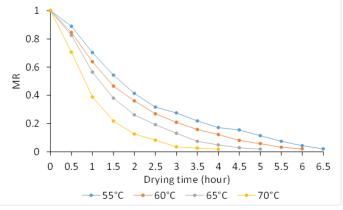


Figure 1. Effect of air temperature and drying time on the moisture ratio of roselle seeds.

### 3.2 Mathematical modelling

The moisture content of samples at different temperatures was converted to the more useful moisture ratio expression, and curve fitting computations with drying time were performed with the five drying models. The results of the statistical analyses undertaken on these models for drying were given in Table 2. The models were evaluated based on Root Mean Square Error (RMSE), coefficient of determination ( $\mathbb{R}^2$ ) and Chi-square ( $\chi^2$ ). All equations gave consistently high  $\mathbb{R}^2$  values in the range of 0.987 to 0. 999. This indicated that all equations could satisfactorily describe the thin layer drying rates of roselle seeds. RMSE ranged from 0.00023 to 0.0388, Chi-square ranged from 0.00025 to 0.0019. Among five thin layer drying models, the Page

Table 2. Modeling the drying process	of roselle seeds u	sing thin-laver	drving at o	different temperatures
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. Modeling	the drying process of re-	oselle seeds usin	g thin-layer	drying at di	fferent temper
	Models	Constants	$\mathbf{R}^2$	$\chi^2$	RMSE
	Newton	k: 0.42857	0.989	0.00134	0.03277
	Logarithmic	k: 0.398964,			
		a: 1.08576	0.994	0.00094	0.02559
		c: -0.0499			
_	Henderson and Pabis	k: 0.450142,	0.993	0.00107	0.02826
55°C		a: 1.0565	0.370	0.00107	0.02020
	Diffusion approach	k: 0.42877,	0.000	0.0019	0.035628
		a: 0.776239	0.989		
		b: 0.998013			
	Page	k: 0.366627,	0.996	0.0007	0.02181
		n: 1.15868		0.00005	0.02752
	Newton	k: 0.511946	0.993	0.00095	0.02753
	Logarithmic	k: 0.472999, a: 1.07563	0.997	0.00051	
			0.997		
	Henderson and Pabis	c: -0.04732 k: 0.5330,			0.02384
60°C		a: 1.0427	0.995	0.00076	
00 C	Diffusion approach	k: 0.512313,			
		a: 0.960	0.993	0.00114	0.03016
		b: 0.99064			
	Page	k: 0.44794,	0.000	0.00005	0.01005
		n: 1.15998	0.999	0.00025	0.01287
	Newton	k: 0.639727	0.987	0.00188	0.0388
		k: 0.579614,			
	Logarithmic	a: 1.09597	0.994	0.00127	0.0298
		c: -0.059304			
	Henderson and Pabis	k: 0.670619,	0.99	0.00189	0.03537
65°C		a: 1.0535	0.99	0.00109	0.05557
		k: 0.427782,	0.000	0.00100	0.001.41
	Diffusion approach	a: 2.09803	0.993	0.00109	0.03141
		b: 0.6983			
	Page	k: 0.549965,	0.998	0.00025	0.01577
	-	n: 1.24767		0.00122	
	Newton	k: 0.944799	0.991	0.00133	0.0333
	Logarithmic	k: 0.897052,	0.994	0.00148	0.03218
		a: 1.0548 c: -0.02984			
		k: 0.971314,		0.00153	0.03306
70°C	Henderson and Pabis	a: 1.03119	0.992		
70 C		k: 0.60,			
	Diffusion approach	a: -6.84242	0.994	0.00122	0.03154
	······································	b: 1.05726	****		
		k: 0.893574,	0.998	0.00033	0.00022
	Page	n: 1.23401			0.00023

model obtained the highest R<sup>2</sup> values of 0.996, 0.999, 0.998 and 0.998 at 55, 60, 65 and 70°C, respectively. Similarly, the lowest RMSE values were obtained in the Page model over the specified temperature range. Thus, this model may be assumed to present the thin-layer drying behaviour of the roselle seeds. These obtained results were similar to the work of some previous studies. da Silva et al. (2014) reported that the Page model is the best in describing the thin-layer drying characteristics of whole bananas at the temperature range of 40 to 70°C. Dzisi et al. (2012) studied the thin-layer modelling of Tetraploid Plantain using hot-air dryer and found that the Page model also satisfactorily described the drying behaviour of the slices at the temperature

range 50 to 80°C. Doymaz (2012) also stated the suitability of the Page model to fit the experimental thin layer drying data of persimmon slices in comparison with other empirical models at 50, 60 and 70°C. Oduola and Oforkansi (2016) reported that the Page model had the least average chi-square  $\chi^2$  value and highest coefficient of determination (R<sup>2</sup>) value of 0.0008 and 0.9922, respectively. Therefore, the Page model best described the drying characteristics of the plantain sample within the 40 to 70°C temperature range. Most recently, Thuy et al. (2021) found that the Page model sufficiently described thin-layer drying of Butterfly pea flowers at 55 to 70°C and Tai et al. (2021) also found that this model was successfully applied to describe the drying characteristics of "Xiem" banana peel. It was observed that roselle seeds can best be dried at 70°C in the shortest time. Comparisons of the predicted values of the Page model with the experimental data were shown in Figure 2. It could be seen that the Page model showed good fits to the experimental values. Besides, the results of the linear regression analysis showed that the data values fluctuated near the straight line with a slope of  $45^{\circ}$ , proving the suitability of the Page model in describing the drying process of roselle seeds, typically the data obtained at a temperature of 70°C with an R<sup>2</sup> value of 0.999 (Figure 3).

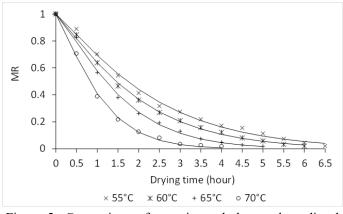


Figure 2. Comparison of experimental data and predicted moisture ratio using Page model for dried roselle seeds.

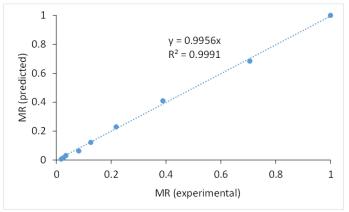


Figure 3. Correlation between the experimentally determined MR values and the estimated MR values for dried roselle seeds using the Page model at  $70^{\circ}$ C.

# 3.3 The effective diffusivity coefficient and activation energy

The effective diffusivity coefficient of dried roselle seeds at 55 to 70°C were varied in the range of  $5.426 \times 10^{-10}$  to  $1.074 \times 10^{-9}$  m<sup>2</sup>/s (Table 3). It was observed that the values of D<sub>eff</sub> increased significantly with increasing temperature. Drying at 70°C gave the highest D<sub>eff</sub> value. The D<sub>eff</sub> values obtained from this study were found to agree with the study of Rasouli *et al.* (2011) for garlic slices at air drying temperatures of 50, 60 and 70°C with the calculated moisture diffusivity coefficient varied from 2.524×10<sup>-10</sup> to 7.566×10<sup>-10</sup>m<sup>2</sup>/s. Tzempelikos *et al.* (2014) studied the effect of the airdrying conditions on the convective drying of Quinces and showed that the values of the effective moisture diffusivity ( $D_{eff}$ ) were obtained between  $2.67 \times 10^{-10}$  and  $8.17 \times 10^{-10}$  m<sup>2</sup> s<sup>-1</sup> in the range of drying temperature of 40-60°C.

Table 3. The moisture diffusivity of roselle seeds at different drying temperatures

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Temperature	The effective diffusivity coefficient	
(°C)	$(m^2/s)$	
55	5.426×10 <sup>-10</sup>	
60	6.390×10 <sup>-10</sup>	
65	$8.240 \times 10^{-10}$	
70	$1.074 \times 10^{-9}$	

However, the D<sub>eff</sub> values obtained from our study were higher than those published by Alara *et al.* (2017) in studying *Vernonia amygdalina* leaves drying, the effective diffusivities for the three air temperatures (40, 50 and 60°C) ranged from  $4.55 \times 10^{-12}$  to  $5.48 \times 10^{-12}$  m<sup>2</sup>/s. Thuy *et al.* (2021) also reported that the effective diffusivity values of dried Butterfly pea flowers at 55 to 70°C were varied in the range of  $2.392 \times 10^{-12}$  to 7.756×10<sup>-12</sup> m<sup>2</sup>/s).

To obtain the effect of temperature on the effective diffusivity, the values of ln ( $D_{eff}$ ) versus 1/T were presented in Figure 4. The plot was found to be a straight line over the temperature ranges investigated, indicating Arrhenius dependency. The activation energy was calculated from the slope of the straight line and it was found to be 43.08 kJ/mol. This value is similar to or slightly higher than the values calculated from the study of Rasouli *et al.* (2011) and Tzempelikos *et al.* (2014), with published E<sub>a</sub> values of 30.42 to 36.365 kJ/mol for garlic slices at 50-70°C and 36.99 to 42.59 kJ/mol for quince slices at 40-60°C, respectively.

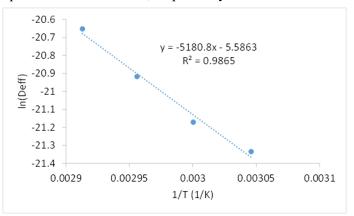


Figure 4. Influence of air temperature on effective diffusivity of dried roselle seeds.

### 4. Conclusion

Drying at 70°C was effective faster than other temperatures, earlier finishing time (4 hrs) and had low product moisture, very favourable for the storage of roselle seeds after collection. A nonlinear regression

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analysis was performed, indicating that Page's thin-layer drying model is best fitted to the experimental results. The values of  $D_{eff}$  were estimated, showing that temperature increases effective moisture diffusivity. The results obtained herein can provide some new theoretical guidelines for the development of the drying model. In addition, to exploit the seeds which contain high-value health-promoting components, their possible application in new food product development should be studied.

# **Conflict of interest**

The authors declare no conflict of interest.

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