MOISTURE SORPTION ISOTHERMS AND THIN-LAYER DRYING OF RICE CRACKERS

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Abstract

In this study, the equilibrium moisture contents of rice cracker samples were experimentally obtained using the gravimetric method at temperatures of 50°C, 60°C, and 70°C, with water activity (aw) ranging from 0.017 to 0.968. The sorption isotherm curves of rice crackers were sigmoidal in shape and decreased with increased temperature at constant relative humidity. Five selected isotherm models were tested to fit the experimental isotherm data of rice crackers. The results showed that the Modified Oswin model fitted the best to the experimental data of rice crackers. Furthermore, thin-layer drying of rice crackers was conducted under controlled conditions of temperature using a convective air dryer. The rice crackers, initially having a moisture content of 85% (d.b.), were dried at temperatures of 50°C, 60°C, and 70°C until their final moisture content reached 6% (d.b.). Besides the effects of drying air temperature and velocity, the Modified Henderson and Pabis model was revealed to be the best, followed by the Two-term model.

Keywords: sorption isotherm, thin-layer drying, rice crackers

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Received: 6 April 2024; Revised: 18 August 2024; Accepted: 20 September 2024

DOI: https://doi.org/10.14456/lsej.2024.23

Introduction

Moisture sorption isotherms describe the relationship between a product's equilibrium moisture content and the relative humidity of the surrounding air at a constant temperature. When drying agricultural produce, it is essential to understand the equilibrium moisture content of the product. In general, the equilibrium moisture content of agricultural products depends on the temperature and relative humidity of the surrounding air (Kaymak & Gedik, 2004; Janjai et al., 2006). Most agricultural products can transfer moisture to the ambient air. This transfer continues until the moisture content decreases to a certain level, at which point it stops. This level of humidity is known as the equilibrium moisture content. A graph that shows the relationship between the relative humidity of the air and the moisture content of a product at a constant temperature is called a sorption isotherm. This data is crucial for predicting shelf-life, determining packaging and storage characteristics, and calculating moisture exchanges during storage of rice crackers. Despite limited research on the sorption isotherm of rice crackers, thin-layer drying modeling is commonly used to understand their drying characteristics (Akgun & Doymaz, 2005; Doymaz, 2009; Midilli et al., 2002; Wang & Singh, 2001).

This study aims to experimentally determine the sorption isotherms of rice crackers, fit an isotherm model to the data, and evaluate their thin-layer drying characteristics to ensure high-quality dried products.

Material and Method

Determination of the sorption isotherm of rice crackers

The equilibrium moisture contents of rice crackers were determined experimentally using the gravimetric method. A sample box, designed as an airtight plastic container, contained a saturated salt solution to maintain a constant relative humidity inside. The samples were placed in perforated containers. These containers were then positioned on perforated plastic supports located just above the salt solution. Sorption isotherm determinations were conducted at temperatures of 50°C, 60°C, and 70°C, with relative humidity ranging from 11% to 97%. The relative humidity values were controlled using various saturated salt solutions as outlined in (Janjai et al., 2007). The final moisture contents of the products were determined using the standard oven method, with a temperature of 103°C maintained for 24 hours.

Selection of the sorption isotherm models

Five isotherm models were tested to fit the sorption isotherm of rice crackers. These models were selected based on their effectiveness in describing isotherms of various food materials. The details of these models are provided in Table 1.

Table 1 Selected isotherm models (Janjai et al., 2007)

No	Model	Mathematical expression
1	Day and Nelson Equation	$a_w = 1\text{-exp}\left[-b_0 T^{b_1} M_e^{b_2 T^{b_3}}\right]$
2	Modified Halsey Equation	$a_w = \exp\left[-\dfrac{\exp(b_0+b_1 T)}{M_e^{b_2}}\right]$
3	Modified Chung-Pfost Equation	$a_w = \exp\left[\dfrac{-b_0}{T+b_1}\exp(-b_2 M_e)\right]$
4	Modified Oswin Equation	$a_w = \dfrac{1}{1+\left[\dfrac{b_0+b_1 T}{M_e}\right]^{b_2}}$
5	Kaleem Ullah Equation	$a_w = b_0\text{-}b_1\exp\left[-b_2 T M_e^{b_3}\right]$

The accuracy of the models in predicting equilibrium moisture content for given values of relative humidity and temperature can be determined by coefficient of determination (R^2) and root mean square error (RMSE). For the best fit, the R^2 value should be high and the lowest RMSE. RMSE and R^2 are defined as:

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

$$R^2 = 1\text{-}\frac{\text{Residual sum of squres}}{\text{Corrected total sum of squares}} \quad (2)$$

where $Me_{exp,i}$ is equilibrium moisture content from experiment (%, db), $Me_{pre,i}$ is equilibrium moisture content from prediction (%, db), N is number of data to be consider.

Drying experiments

The initial moisture content of rice crackers about 85% (db). The rice crackers, with a diameter of 4.5 cm and a thickness of 0.75 cm, were placed in a thin layer on a tray in a laboratory dryer and dried under controlled temperature conditions, as shown in Figure 1.

Figure 1 The rice crackers were placed on a tray in a laboratory dryer.

The rice crackers were dried at the temperature of 50 °C, 60 °C and 70 °C with the air speed of 1 ms^{-1}. A schematic diagram of this laboratory dryer is shown in Figure 2. It consists of a blower, heaters, drying chamber, and instruments for measurement. The airflow rate was adjusted by the fan speed control. The heating system consisted of an electric heater placed inside the duct. The drying chamber temperature was adjusted by the heater power control. The homogeneity of air temperature and air flow was checked by using thermocouples and a hot wire anemometer. The moisture content during drying was estimated from the weight of the product samples and the estimated dried solid mass of the samples. At the end of the experimental drying, the exact dry solid mass of the product samples was determined by the oven method (103 °C for 24 hours, accuracy ± 0.5%). The thin-layer drying tests were conducted in the temperature range of 50-70 °C and nine sets of experiments were conducted for the rice crackers.

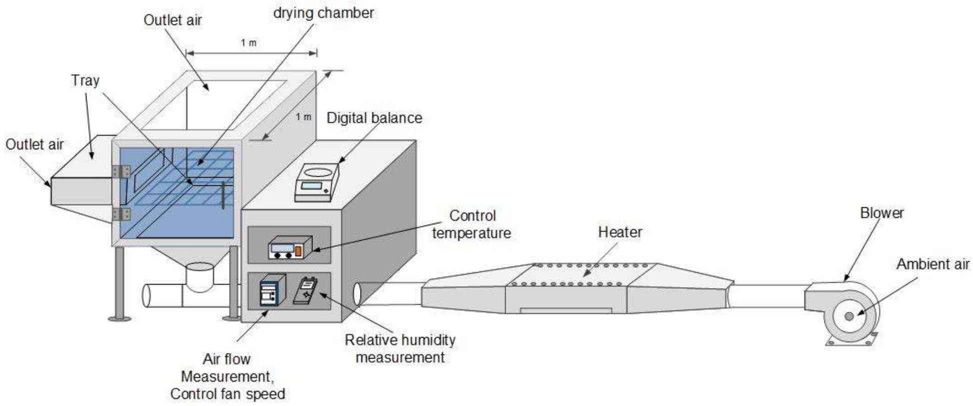

Figure 2 The schematic diagram shows the laboratory dryer.

Mathematical modeling

There are three approaches to the modeling of thin-layer drying of products. These are: (a) theoretical approach, (b) semi-theoretical approach and (c) empirical approach. A theoretical equation gives a better understanding of the transport processes but an empirical equation gives a better fit to the experimental data without any understanding of the transport processes involved. The semi-theoretical equation gives some understanding of the transport processes. Thin-layer drying models of experimental data of the rice crackers are expressed in the form of moisture content ratio of samples during drying, and it is expressed as:

$$MR = \left[\frac{M - M_e}{M_0 - M_e}\right] \tag{3}$$

Where MR is the dimensionless moisture content or moisture ratio; and M, M_0 and M_e are the moisture content at any given time, the initial moisture content and the equilibrium moisture content, respectively.

In general, an agricultural moist product is composed of water and dried solid mass. The moisture content (M) of the product in dry basis (% db.) can be calculated from the following equation:

$$M = \frac{m - m_{solid}}{m_{solid}} \times 100 \tag{4}$$

Where m is mass of the product and m_{solid} is mass of dried solid mass of the product. m can be obtained by using a digital balance. In order to obtain dried solid mass (m_{solid}), the water in the product must be totally removed by drying the product in an oven at the temperature of 103 °C for 24 hours. To select a suitable model for describing the drying process of rice crackers, seven different thin-layer drying models were selected to fit the thin-layer experimental data of rice crackers. The selected thin-layer drying models are presented in Table 2. The models were fitted to the experimental data by direct least square. The coefficient of determination (R^2) was one of the main criteria for selecting the best equation. In addition to R^2, the goodness of fit was determined by root mean square error (RMSE).

Table 2 The 7 selected thin-layer drying models.

No.	Model equation	Name of the model
1	MR=exp(-kt)	Newton (Mujumdar, 1987)
2	MR=exp(-kt^n)	Page (Diamante & Munro, 1993)
3	MR=1+at+bt^2	Wang and Singh (Whith et al., 1978)
4	MR=a exp(-kt)	Handerson and Pabis (Zhang & Litchfield, 1991)
5	MR=a exp(-kt)+c	Logarithmic (Yangcioglu et al.,1999)
6	MR=a exp(-kt)+b exp(-gt)	Two terms (Henderson, 1978)
7	MR=exp(-kt)+b exp(-gt)+c exp(-pt)	Modifier Handerson and Pabis (Karathanos, 1999)

Results

Sorption isotherm

Results from the experiment to find out sorption isotherm of rice crackers at 50°C, 60°C and 70°C and water activity (a_w) is in the range of 0.017 to 0.968 were shown in Figure 3. It could be seen that the isotherm curve was in the shape of sigmoid. The constant in the form of equation coefficient, the accuracy of the simulation model (RMSE), the difference between the prediction result and the experiment result (R^2) for sorption isotherm of rice crackers were shown in Table 3. According to the test through isotherm models to predict, the Modified Oswin model was the most suitable method to predict the experiment result for isotherm of rice crackers at 50 °C, 60 °C and 70 °C.

However, it must be mentioned that the goodness of fit of a sorption model to experimental data does not describe the nature of the sorption process, it only reflects on the mathematical quality of the model (Samapundo et al., 2007).

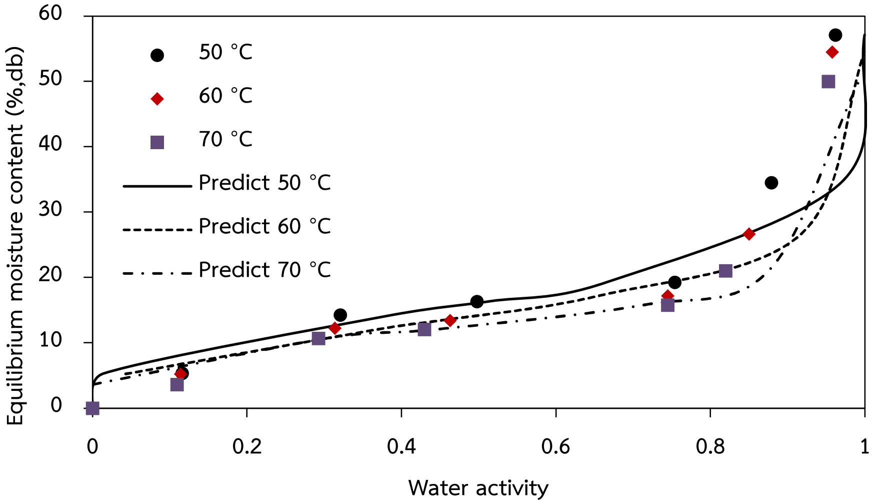

Figure 3 Predicted (Modified Oswin model) and measured sorption isotherms of rice crackers at 50 °C, 60 °C and 70 °C.

Table 3 The coefficients of the selected models, standard error of estimate (RMSE) and the coefficient of determination (R^2) for rice crackers.

Model	Temperature (°C)	Coefficients				RMSE	R^2
		b_0	b_1	b_2	b_3		
Day and Nelson	50, 60, 70	0.170093	-0.90634	0.421197	0.293988	3.88	0.962
Modified Halsey	50, 60, 70	7.030747	-0.01423	1.810505	-	4.55	0.961
Modified Chung-Pfost	50, 60, 70	301.5683	45.4092	0.0364	-	6.12	0.965
Modified Oswin	50, 60, 70	56.36232	-0.31521	2.06275	-	1.18	0.967
Kaleem Ullah	50, 60, 70	-7.55323	-7.61228	-0.00023	0.41139	1.31	0.726

Five models of the isotherm of rice crackers; the Modified Oswin model fitted the best to the experimental data of rice crackers. The agreement between the best-fitted models and experimental data was excellent. For simplicity and consistency, effect of temperature i.e. equilibrium moisture content decreases with the increase of moisture content, Modified Oswin was selected for use in this simulation. Another advantage of

Modified Oswin is that equilibrium moisture content can be calculated directly as a function of temperature and water activity using only one set of coefficients, thus facilitating the calculation. This equilibrium moisture content model is written as:

$$a_w = \frac{1}{1+\left(\frac{56.36232+(-0.31521\times T)}{M_e}\right)^{2.06275}} \quad (5)$$

where b_0 = 56.36232, b_1 = -0.31521, b_2 = 2.06275 is a function of temperature and humidity. T is temperature (°C), a_w is water activity (decimal) and M_e is equilibrium moisture content (%, db). The water activity is equal to the relative humidity (%) divided by 100.

Drying characteristics of rice crackers

Figure 4 illustrates the changes in moisture content over time for rice crackers at different drying air temperatures. The final moisture content of samples dried under different conditions is approximately 6% on a dry basis (db). Figure 4 shows the comparison of predicted and experimental data for thin-layer drying of rice crackers using the modified Henderson and Pabis model.

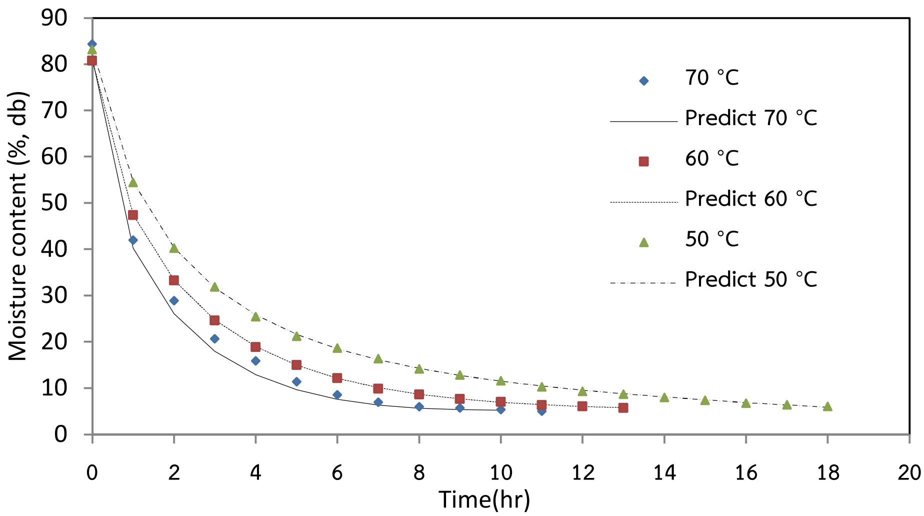

Figure 4 Predicted and observed moisture content of rice crackers using modified Handerson and Pabis model at the temperatures of 50°C, 60°C and 70°C.

Table 4 Parameter value, coefficient of determination (R^2) and root mean square error (RMSE) value of the different models for rice crackers.

Models	T(°C)	k	a	b	c	n	g	p	R^2	RMSE (%)
	50	0.26108							0.9207	6.55
Newton	60	0.36746							0.9523	5.53
	70	0.52324							0.9646	5.10
	50	0.47485				0.62040			0.9470	1.26
Page	60	0.56429				0.65495			0.9971	1.22
	70	0.72440				0.64643			0.9977	1.12
Wang and	50			0.00640					0.7696	11.17
Singh	60			0.01227					0.8311	10.41
	70			0.02125					0.8374	10.94
Henderson	50	0.22136	0.87634						0.9412	5.64
and Pabis	60	0.33366	0.92200						0.9604	5.04
	70	0.49351	0.95003						0.9679	4.86
	50	0.36254	0.86174		0.10009				0.9904	2.28
Logarithmic	60	0.48714	0.89043		0.08638				0.9946	1.85
	70	0.67599	0.90988		0.07585				0.9952	1.88
	50	0.09780	0.4398	0.61779			0.64110		0.9994	0.55
Two-term	60	0.75606	0.6857	0.33826			0.13515		0.9988	0.89
	70	0.19046	0.4023	0.67572			1.03010		0.9987	0.99
Modified	50	0.40186	0.53196	0.55161	0.19357		0.07556	1.46045	0.9999	0.22
Henderson	60	1.88572	0.24452	0.27444	0.07324		0.35629	0.00912	0.9999	0.09
and Pabis	70	-0.0837	0.02350	0.68224	0.30723		0.41837	2.33277	0.9999	0.16

Mathematical modeling of thin-layer drying

Seven thin-layer drying model (Table 2) were fitted to the experimental data of moisture ratio of rice crackers dried at different temperatures and relative humidity. The parameter values, R^2 and RMSE, are also shown in Table 4. The Modified Handerson and Pabis model was found to be the best, followed by the Two term model. The value of R^2 of the Modified Handerson and Pabis model was 0.9999, indicating good fit and RMSE was 0.16-0.22. Empirical expressions were developed for the drying parameters of the Modified Handerson and Pabis model and the drying parameters were found to be a function of drying air temperature (T in °C) and relative humidity (rh in %):

The parameter values for rice crackers.

$$k=-48.294001+1.686829T-0.035136rh+0.003004Trh-0.014543T^2-0.003075rh^2 \quad (6)$$

$$a=-4.764590+0.237951T-0.205002rh+0.001673Trh-0.002313T^2+0.002800rh^2 \quad (7)$$

$$b=-1.897929+0.013931T+0.229736rh-0.002430Trh+0.00273T^2-0.002364rh^2 \quad (8)$$

$$c=7.658853-0.251754T-0.024804rh+0.000756Trh+0.002039T^2-0.000433rh^2 \quad (9)$$

$$g=-1.635049+0.029858+0.088287rh-0.001052Trh-0.000010T^2-0.000883rh^2 \quad (10)$$

$$p=-219.148655+7.636277T-0.940576rh-0.001183Trh-0.062994T^2+0.033341rh^2 \quad (11)$$

Discussion

The equilibrium moisture content decreased when the temperature increased at all levels of relative humidity. This could be explained as follows. The increasing temperature made water molecule increase so much kinetic energy that it could break attraction forces between molecules. The water in the product would evaporate to surrounding air more than in the low temperature. The equilibrium moisture content, therefore, decreases when the drying temperature increases. Besides, when the temperature is constant, the equilibrium moisture content will increase when the relative humidity in the surrounding air increases. This is due to the fact that high relative humidity in the surrounding air reduces the vapor pressure difference between the surrounding air and the product, leading to minimal water evaporation from the product surface to the surrounding air. The isotherm curves exhibit similar patterns at different temperatures, with equilibrium moisture content values decreasing as the temperature increases at a constant relative humidity. This suggests that rice crackers become less hygroscopic as the temperature rises. This phenomenon can be explained by the increase in kinetic energy associated with the water molecules in the rice crackers as the temperature increases. The increased kinetic energy reduces the attractive forces between the water molecules, allowing them to escape more easily, which in turn decreases the equilibrium moisture content. Similar trends have been reported in several studies on plant and food materials (Shivhare et al., 2004; Mohamed et al., 2005) shown in Figure 3. Additionally, it takes longer to reach equilibrium status under these conditions. Furthermore, the drying rate increases with higher air temperatures, leading to a shorter time required to reach the final moisture content. Therefore, the drying air temperature significantly influences the drying process of rice crackers.

Conclusions

The equilibrium moisture contents of rice crackers were experimentally determined using the gravimetric method at temperature levels of 50°C, 60°C, and 70°C, with water activity (aw) ranging from 0.017 to 0.968. All the isotherms exhibited a sigmoidal shape, with the equilibrium moisture content decreasing as the temperature increased at constant water activity. Five sorption isotherm models were applied to fit the experimental data of rice crackers, and among them, the modified Oswin model showed the best fit at the temperature levels of 50°C, 60°C, and 70°C. This model is recommended for use in the drying process of rice crackers.

The thin-layer drying of rice crackers, finding that the drying rate increases with higher air temperatures. The entire drying process occurred in the falling rate period, and no constant rate period was observed. The rice crackers, initially having a moisture content of 85% (d.b.), were dried at temperatures of 50°C, 60°C, and 70°C until their final moisture content reached 6% (d.b.). Seven thin-layer drying models were applied to the experimental data of rice crackers to describe their drying characteristics. The drying parameters of the Modified Henderson and Pabis model were found to be a function of the drying air temperature. Among the models tested, the Modified Henderson and Pabis model performed the best, followed by the Two-term model. This suggests that the Modified Henderson and Pabis model is suitable for assessing the drying behavior of rice crackers and for simulating and optimizing the dryer for efficient operation.

Acknowledgements

The author would like to thank Program of Physics, Department of Science, Faculty of Science and Technology. Thanks also go to Professor Dr. Serm Janjai, Department of Physics, Faculty of Science, Silapakorn University.

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