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Model for Thin Layer Drying of Lemongrass (*Cymbopogon citratus*) by Hot Air

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Abstract: Lemongrass is a plant that contains aromatic compounds (myrcene and limonene), powerful deodorants, and antimicrobial compounds (citral and geraniol). Identifying a suitable drying model for the material is crucial for establishing an initial step for the development of dried products. Convection drying is a commonly used drying method that could extend the shelf life of the product. In this study, a suitable kinetic model for the drying process was determined by fitting moisture data corresponding to four different temperature levels: 50, 55, 60 and 65 °C. In addition, the effect of drying temperature on the moisture removal rate, the effective diffusion coefficient and activation energy were also estimated. The results showed that time for moisture removal increases proportionally with the air-drying temperature, and that the Weibull model is the most suitable model for describing the drying process. The effective diffusion coefficient ranges from 7.64×10^{-11} m^2/s to 1.48×10^{-10} m^2/s and the activation energy was 38.34 kJ/mol. The activation energy for lemongrass evaporation is relatively high, suggesting that more energy is needed to separate moisture from the material by drying.

Keywords: lemongrass; *Cymbopogon citratus*; convection drying; mathematical modeling; moisture diffusivity; activation energy

1. Introduction

Lemongrass (*Cymbopogon citratus*), also known as oil grass, silky heads, or citronella grass, is a popular annual crop for spices and medicinal herbs. The species is an herbaceous plant with long, thin leaves, and belongs to the Cymbopogon genus that is geographically distributed over various regions in India, America, Africa, Australia and Europe [1]. The use of lemongrass is diverse. At the family scale, lemongrass in both fresh and dried forms, is used as a spice in Vietnamese and Thai cooking, and for treatment of the common cold [2]. In Chinese and Indian medicine, the plant has been widely utilized as a tranquilizer and anti-inflammatory medicine. Tea made from lemongrass is also commonly consumed in Brazil, Cuba, and Argentina as a treatment for catarrh, rheumatism, and sore throat [3]. At larger scale, lemongrass is often cultivated and harvested to produce citronella oil and citronella-derived products such as mosquito repellant, soap, and perfume. The broad variety of uses of lemongrass is due to its valuable and numerous biological properties, which have been extensively studied and documented, including antifungal, antibacterial, antioxidant, anti-carcinogenic, and anti-rheumatic activities [3,4].

Lemongrass in general, and lemongrass oil in particular, is known for its abundant citral component, which is composed of geranial and neral. The content of citral in lemongrass essential oil varies from 40 to 82%, and is considered as an important quality indicator for utilization in medicine, food, herbicides, and cosmetics [5]. Lemongrass is also rich in aromatic compounds, which have been demonstrated to exhibit anticancer activity (myrcene and limonene) and pain-relieving properties (myrcene and limonene). Regarding antimicrobial properties, citral, mycrene, geranial, and gereniol—which are found in lemongrass in large proportions—have been shown to inhibit *Desulfovibrio alaskensis* (gram-negative bacteria), *Campylobacter jejuni* (enteritis-causing bacteria), *Escherichia coli* O157 (diarrhea-causing bacteria), and *Listeria monocytogenes* [6,7]. Other components with beneficial properties include limonene (antioxidant capacity), β-myrcene (gout prevention), citronellol (anti-fungal properties), and methyl heptentone (anti-diabetes, allergy, and anti-cancer) [6].

Similar to other aromatic plants, the nutritional quality of raw lemongrass cannot be maintained for an extended period of time, highlighting the need for the food industry to find a way to produce low-moisture lemongrass and select a suitable preservation method [8]. Drying, which involves the process of moving water inside the materials onto the surface and removing surface water by evaporation, is a common and efficient preservation technique [9]. Drying could prevent multiplication of spoilage-causing micro-organisms and minimize the occurrence of chemical reactions inside the material, in turn prolonging the shelf-life of the product and resulting in higher economic value than direct use of raw materials. Among many feasible methods of separating moisture from materials (such as convection drying, vacuum drying, freeze drying, and fluidized bed drying), thin layer hot air convection drying has proven to be a simple and cost-effective method [10]. However, to develop an efficient drying system for a particular product, the determination of suitable drying models for raw materials is required. Accurate modeling of material behavior during the drying process contributes to the development of dried products by predicting the drying process of materials based on the moisture balance of the material, from which improvements to existing drying system can be made [10].

Although lemongrass has been extensively studied with regards to its biological activities and chemical composition, literature concerning drying kinetics of lemongrass is still limited. To the best of our knowledge, two notable attempts have been made to model lemongrass behavior in drying processes [11,12]. The study of Coradi et al. (2014) suggested the appropriateness of the two-term model in describing the drying kinetics of lemongrass, demonstrated by a low value of the average estimated error (SE) and average relative error (P) [11]. However, only four typical models were taken into account in their study: modified Page, logarithmic, two-term and Midilli models. On the contrary, Simha (2016) fitted experimental data to an extensive pool of ten drying models, showing that the Midilli model is the most suitable drying kinetics model for both lemongrass and *Adathoda vasica* leaves [12]. However, the drying method of choice in this study was microwave drying, which lacks applicability and is energy-inefficient at larger scale.

Given the scarcity of studies on lemongrass drying kinetics and the significance of the thin layer convection-drying technique, this study aimed to determine the kinetic profile that characterizes convective drying of lemongrass material. In addition, the effective moisture diffusion coefficient, activation energy of the material, and other important parameters resulting from the present study can act as a precursor for further investigation regarding the mass transfer and moisture removal occurring during the drying process under various drying conditions.

2. Materials and Methods

2.1. Materials and Methods

2.1.1. Materials

Lemongrass (*Cymbopogon citratus*) was purchased at Vuon Lai Market, District 12, Ho Chi Minh City, Vietnam. Harvested lemongrass was planted within 10 to 12 months. The initial moisture content of the material was 6.40 ± 0.15 (g water/g dry matter). Before drying, lemongrass was pre-treated

by washing and cut into uniformly sized samples with a length of 38.95 ± 1.35 cm and a thickness of 1.46 ± 0.14 mm (Figure 1). The moisture content was determined using the oven method [13].



Figure 1. The lemongrass material.

2.1.2. Drying Equipment

A pilot-scale hot air convection dryer was used to dry lemongrass slices. The dryer consisted of two drying rooms, in which each room contained 10 wire mesh trays. The drying tray size was 80 × 80 × 3 cm and the mesh size was 1 × 1 cm (Figure 2). Moist air from the external environment at 30.5 ± 1.0 °C and 70% RH was fed to the dryer by exhaust fans where it was heated to a maximum temperature of 65 °C. The heated air then came into contact with materials, absorbing moisture and exiting the dryer.

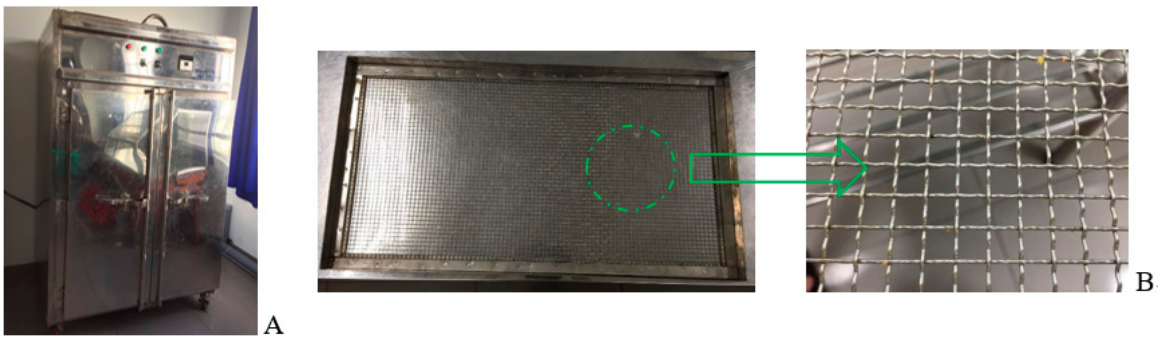


Figure 2. Convection dryer (**A**) and tray (**B**) used in this study.

2.1.3. Drying Procedure

For each sample, a thin layer (1000 g) was put in the tray dryer. In this study, the air-drying temperature was set at 50, 55, 60 and 65 °C. The sample weight was recorded during drying at 10-min interval. Experimental data was used to evaluate mathematical models of drying curve as well as calculate the effective moisture diffusivity and activation energy.

A drying temperature range of 50–65 °C is generally feasible for a wide number of medicinal plants [14] and was selected in this study for a number of reasons. First, it is suggested that, to obtain the highest essential oil content, drying of lemongrass should be conducted in an oven at 45 °C for 7 h [15]. In another study, it is suggested that temperature of drying should range from 50 to 70 °C to achieve maximum oil retention and that a temperature of below 30 °C was not advisable due to

the promotion of fungi [16].Second, citral content, which determines the quality of the lemongrass products, was shown to be insensitive to thermal treatment in the range of 50–65 °C. Another study attempting drying of lemon myrtle leaves suggested that the optimal drying temperature for maximum citral retention was 50 °C [17] and that slightly higher drying temperature was not detrimental to citral content. This is due to the protective effects of the partially dried surface layers formed by high temperature, limiting the mass transfer and the diffusion of volatile components to the surface. This result is supported by Rocha et al., (2000) who stated that both the oil yield and composition of citronella (*Cymbopogon winterianus*) were at optimal levels at a drying temperature of 60 °C [18].

2.2. Determination of Mathematical Components

2.2.1. Moisture Content

Moisture content (g water/g dry matter) was calculated by the following equation [19].

$$M = \frac{m_w}{m_{dm}}, \quad (1)$$

where M is the moisture content (g water/g dry matter), m_w is the mass of water in sample (g), and m_{dm} is the mass of dry matter in sample (g)

2.2.2. Drying Rate

Drying rate (DR) is defined as the amount of evaporated moisture over time. The drying rate (g water/g dry matter/min) during the process of drying lemongrass was determined using the following equation:

$$DR = \frac{M_t - M_{t+dt}}{dt}, \quad (2)$$

where M_t is the moisture content at t time (g water/g dry matter), M_{t+dt} is the moisture content at t + dt time (g water/g dry matter), and dt is drying time (min).

2.2.3. Mathematical Modeling of Drying Curves

Moisture ratio is defined as follows [20].

$$MR = \frac{M_t - M_e}{M_0 - M_e}, \quad (3)$$

where M_t is the moisture content (g water/g dry matter). The subscripts t, 0, and e denotes time t, initial, and equilibrium, respectively.

To identify the suitable mathematical model for lemongrass drying, the experimental data were fitted to different thin-layer drying models (Table 1).

Evaluation of mathematical models was performed based on coefficient of determination (R^2), root mean square error (RMSE) and chi-squared (x^2). The criteria for a suitable model included high a R^2 value, and low x^2 and RMSE values. The statistical values were defined as follows [21]:

$$\text{The coefficient of determination : } R^2 = 1 - \frac{\sum_{i=1}^{N} \left(MR_{exp,i} - MR_{pre,i}\right)^2}{\sum_{i=1}^{N} \left(\overline{MR_{exp,i}} - MR_{pre,i}\right)^2} \quad (4)$$

$$\text{The Root Mean Square Error : } RMSE = \sqrt{\frac{1}{N}\sum_{i=1}^{N} \left(MR_{exp,i} - MR_{pre,i}\right)^2} \quad (5)$$

$$\text{Chi} - \text{squared}: \ \chi^2 = \frac{\sum_{i=1}^{N} \left(MR_{\text{exp,i}} - MR_{pre,i}\right)^2}{N - Z} \quad (6)$$

where MR_{exp} is the experimental dimensionless moisture ratio, MR_{pre} is the predicted dimensionless moisture ratio, $\overline{MR_{exp}}$ is the mean value of the experimental dimensionless moisture ratio, N is the number of observations, and Z is the number of constants in the mathematical model.

2.2.4. Estimation of the Effective Moisture Diffusivity

To determine the moisture diffusion of a material, the Fick diffusion model was applied as follows [21].

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

where M is the moisture content (g water/g dry matter), D_{eff} is the effective moisture diffusivity (m^2/s), and x is position with the dimensions of length (m).

Crank (1975) has provided a solution that deals with different shapes [22]:

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

where MR is the dimensionless moisture ratio, L is the half-thickness (m), n is the term in series expansion, and t is time (s).

When drying for a long time, the above equation can be reduced to:

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

The effective moisture diffusivity could be described by empirical data using the graph of ln(MR) versus time (t) and the slope of straight line from the plot as $-\pi^2 \frac{D_{\text{eff}}}{4L^2} t$.

2.2.5. Estimation of Activation Energy

The activation energy is calculated by the Arrhenius equation [20,23]:

$$D_{\text{eff}} = D_0 \exp\left(-\frac{E_a}{RT}\right), \quad (10)$$

where E_a is the activation energy (kJ/mol), R is the ideal gas constant (8.3143 kJ/mol), T is the absolute temperature (K), and D_0 is the pre-exponential factor (m^2/s).

2.2.6. Data Analysis

Microsoft Excel software was used to calculate moisture content, moisture ratio, and determine the effective moisture diffusivity and activation energy of the drying process. MATLAB 2014R software was used to find the best model fit and the mathematical model coefficients.

3. Results

3.1. Drying Curves and Drying Rate Curves

Figure 3 shows the change in moisture ratio of lemongrass at different temperatures, from the initial moisture content of 6.40 g water/g dry matter to the constant moisture content. It took 310, 290, 260 and 200 min to completely dry lemongrass at 50, 55, 60 and 65 °C, respectively. The results indicate that higher air-drying temperature resulted in greater slope of the curve and shorter drying time. The explanation for this could be two-fold. First, since moisture removal of the material occurs in

parallel with moisture diffusion from center to surface material and from the surface to environment, higher air-drying temperature reduces relative humidity on the surface material, which in turn promotes surface evaporation in the drying process [23]. Second, increased air-drying temperature also leads to an improved temperature gradient and surface evaporation rate, accelerating moisture diffusion from the center to the surface. These results are consistent with another study, where the decreased drying time was attributed to increases of the air-drying temperature [24].

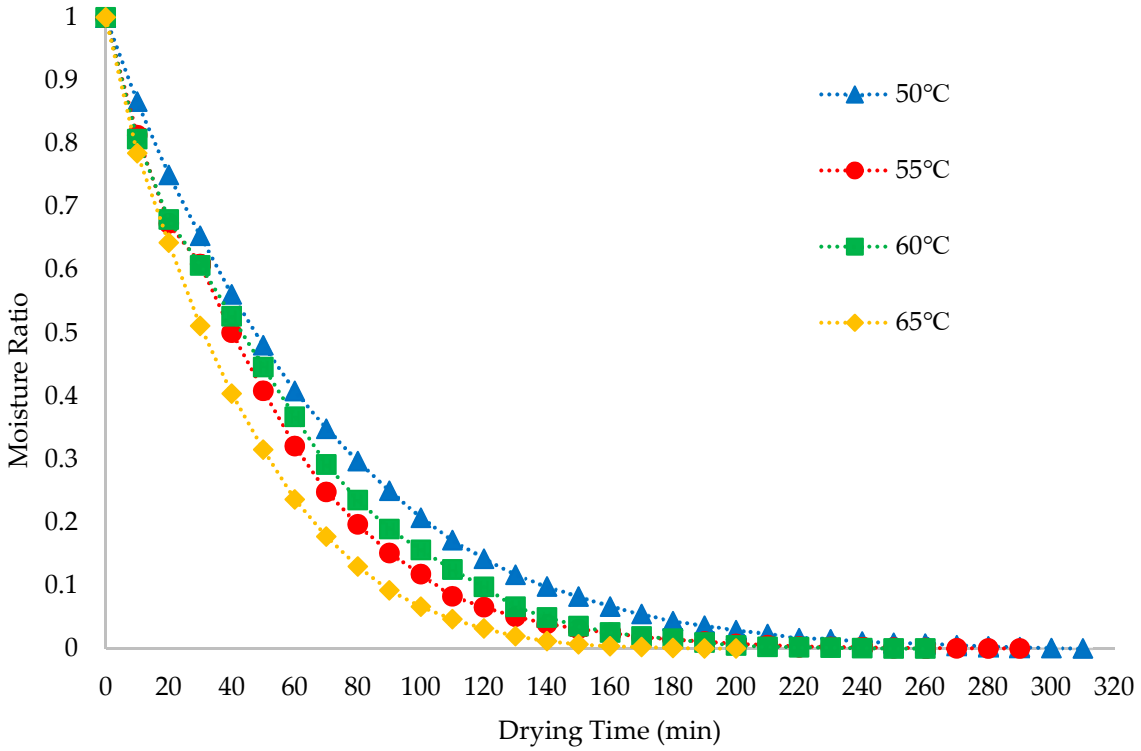


Figure 3. Drying curves of lemongrass slices at different temperature.

Figure 4 shows the drying rate curves of lemongrass slices at different temperatures. Evidently, higher drying temperature was associated with increased drying rate. Since higher temperature induces more heat transfer to the sample, which in turn leads to increased moisture diffusion to the inside and the outside of the materials, moisture removal was accelerated at higher air-drying temperatures. On the other hand, the drying process pattern described in Figure 4 only exhibited two periods—the initial and falling-rate periods. This is different from a standard drying process consisting of three periods: (i) initial period, (ii) constant-rate period, and (iii) falling-rate period. To be specific, the period starting from the initial moisture content to the moisture content of 5 g water/g dry matter coincided with the initial drying period and the period with moisture of higher than 5 g water/g dry matter represented the falling-rate period. The absence of the constant drying rate could be explained by the thin-layer arrangement and the high flow of the drying agent, which quickly accelerates evaporation and circumvents the saturation state of the material. Recent studies have also shown that the constant rate period was absent in the drying processes of fruits and vegetables, since this period often occurs very quickly [25]. At very low moisture content, of less than 0.2 g water/g dry matter, differences between drying rates of the four temperature levels were indistinguishable. This could be mainly due to the lack of water after the removal of free moisture, leading to the diversion of thermal energy into the breaking of bonds instead of heating water molecules. However, since the remaining water molecules were strongly bond to the cellulose fibers, increasing drying temperature from 50 to 65 °C was inadequate to induce a noticeable change in drying rate for lemongrass materials with low moisture.

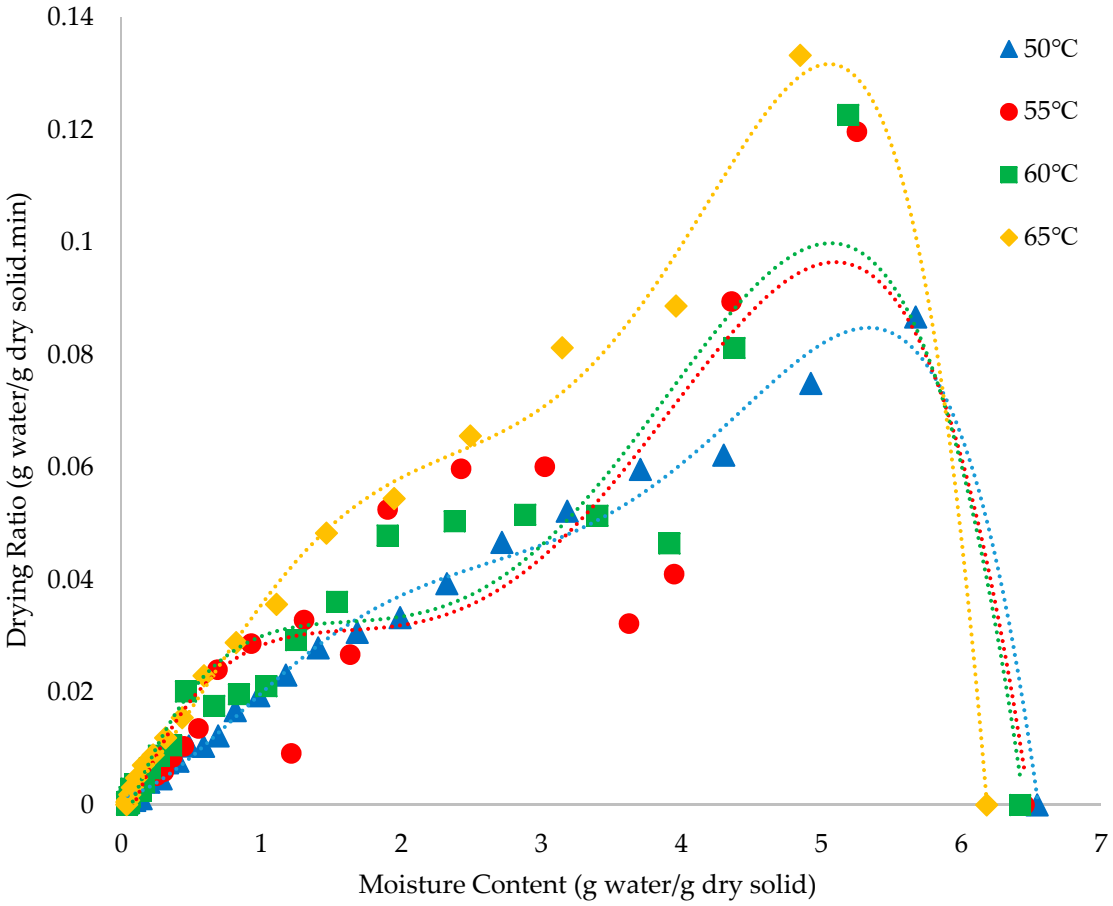


Figure 4. Drying rate curves of lemongrass slices at different temperatures.

3.2. *Mathematical Models of Drying Curves*

Seven common thin-layer drying models (Table 1) were fitted to assess the suitability of experimental data and the results of nonlinear regression analysis are presented in Table 2. The most suitable model to describe lemongrass drying is the model giving highest coefficient of determination (R^2), lowest Root Mean Square Error (RMSE), and lowest chi-square (x^2). From the results of Table 2, all models show that the R^2 values range from 0.93582 to 0.99983 and the RMSE values range from 0.00362 to 0.06887 and x^2 range from 1.50×10^{-5} to 5.08×10^{-5}. Therefore, any of these models of thin layer drying can be used to estimate the change in moisture content over time [26].

Table 1. Mathematical models used to predict the moisture ratios values [25].

No	Model	Equations
1	Page	$MR = \exp(-kt^n)$
2	Henderson and Pabis	$MR = a\exp(-kt)$
3	Midilli	$MR = a\exp(-kt^n) + bt$
4	Logarithmic	$MR = a\exp(-kt) + c$
5	Two-term	$MR = a\exp(-k_1t) - b\exp(-k_2t)$
6	Wang and Singh	$MR = 1 + at + bt^2$
7	Weibull	$MR = a - b\exp(-k_0t^n)$

Table 2. Result of statistical analyses on the modeling of moisture content and drying time.

Temp.	Model	Coefficients				Statistics		
						R^2	RMSE	x^2
50 °C	Page	k = 0.00011	n = 1.10025			0.99959	0.00557	3.30×10^{-5}
55 °C		k = 0.00015	n = 1.07613			0.99505	0.01912	3.92×10^{-4}
60 °C		k = 0.00014	n = 1.09420			0.99555	0.01861	3.74×10^{-4}
65 °C		k = 0.00017	n = 1.10771			0.99865	0.01048	1.21×10^{-4}
50 °C	Henderson and Pabis	a = 1.02592	k = 0.00026			0.99790	0.01266	1.71×10^{-4}
55 °C		a = 1.00482	k = 0.00029			0.99377	0.02147	4.94×10^{-4}
60 °C		a = 1.00910	k = 0.00031			0.99361	0.02229	5.36×10^{-4}
65 °C		a = 1.01880	k = 0.00042			0.99661	0.01660	3.05×10^{-4}
50 °C	Midilli	a = 0.99120	k = 0.00011	n = 1.09672	$b = -4.12 \times 10^{-7}$	0.99980	0.00386	1.65×10^{-5}
55 °C		a = 0.96973	k = 0.00012	n = 1.09701	$b = -6.75 \times 10^{-7}$	0.99607	0.01703	3.22×10^{-4}
60 °C		a = 0.97110	k = 0.00012	n = 1.09938	$b = -1.19 \times 10^{-6}$	0.99704	0.01516	2.59×10^{-4}
65 °C		a = 0.98882	k = 0.00018	n = 1.09637	$b = -1.07 \times 10^{-6}$	0.99917	0.00821	7.86×10^{-5}
50 °C	Logarithm	a = 1.03448	k = 0.00025	c = −0.01985		0.99923	0.00769	6.52×10^{-5}
55 °C		a = 1.01527	k = 0.00027	c = −0.02295		0.99574	0.01775	3.50×10^{-4}
60 °C		a = 1.02510	k = 0.00028	c = −0.03240		0.99682	0.01573	2.70×10^{-4}
65 °C		a = 1.03218	k = 0.00038	c = −0.02517		0.99870	0.01027	1.23×10^{-4}
50 °C	Two-term	a = 0.50040	k_1 = 0.00026	b = 0.49978	k_2 = 0.00026	0.99730	0.01435	2.35×10^{-4}
55 °C		a = 0.55424	k_1 = 0.00029	b = 0.44483	k_2 = 0.00029	0.99373	0.02152	5.34×10^{-4}
60 °C		a = 0.50114	k_1 = 0.00030	b = 0.50058	k_2 = 0.00031	0.99356	0.02238	5.88×10^{-4}
65 °C		a = 0.49822	k_1 = 0.00041	b = 0.50202	k_2 = 0.00041	0.99628	0.01740	3.74×10^{-4}
50 °C	Wang and Singh	a = −0.00016	b = 0.00000			0.94964	0.06197	4.10×10^{-3}
55 °C		a = −0.00017	b = 0.00000			0.93582	0.06887	5.08×10^{-3}
60 °C		a = −0.00019	b = 0.00000			0.95715	0.05773	3.60×10^{-3}
65 °C		a = −0.00025	b = 0.00000			0.95231	0.06229	4.29×10^{-3}
50 °C	Weibull	a = −0.00803	b = −1.00008	k_0 = 0.000117	n = 1.08916	0.99983	0.00362	1.50×10^{-5}
55 °C		a = −0.01311	b = −0.98494	k_0 = 0.000138	n = 1.08109	0.99620	0.01676	3.24×10^{-4}
60 °C		a = −0.02076	b = −0.99429	k_0 = 0.000143	n = 1.07915	0.99725	0.01463	2.51×10^{-4}
65 °C		a = −0.01382	b = −1.00375	k_0 = 0.000199	n = 1.08323	0.99927	0.00772	7.37×10^{-5}

Based on the results of the statistical analysis and the estimated coefficients of the mathematical models shown in Table 2, the Weibull model achieved the highest values of R^2, averaging at 0.99813, the lowest values of x^2, averaging at 0.000166, and lowest RMSE, averaging at 0.010683. This indicates that the model Weibull is the best model to estimate moisture ratio of lemongrass compared to the other models, followed by the Midilli model in which R^2, x^2, and RMSE averaged at 0.99802, 0.000169, and 0.011065 respectively. Since the Weibull model is rarely included in comparison studies of drying kinetics, the present results are in line with results of Onwude et al. (2016), demonstrating that approximately 24% of the literature sources supports the Midilli model; and with Simha et al. (2016), who affirmed the suitability of the Midilli model in microwave drying of *Cymbopogon citratus* [12,25]. From Table 2, it can also be seen that the predictive power of the Midilli model is weaker than that of the Weibull model in terms of R^2, RMSE, and x^2. At each air-drying temperature, the Weibull model exhibited higher R^2, and lower RMSE and x^2 in comparison with the other fitted models. In addition, the Weibull model estimates showed that the drying constant k_0 increased when the temperature increased from 50 to 65 °C. This result is consistent with previous studies, where the Weibull model was suggested to be able to well-describe the drying kinetics of fruits and various vegetables such as garlic, quinces, and persimmon [25].

3.3. Estimation of the Effective Moisture Diffusivity

Figure 5 shows the approximated linear relationship between ln(MR) and drying time (t) at different drying temperatures of 50, 55, 60 and 65 °C. Linear regression was used to calculate the effective moisture diffusion coefficient. The effective moisture diffusion coefficient and the R^2 correlation coefficient correspond to each temperature and were calculated by linear regression of experimental value. ln(MR) (dimensionless) with drying time (t) (second) are presented in Table 3.

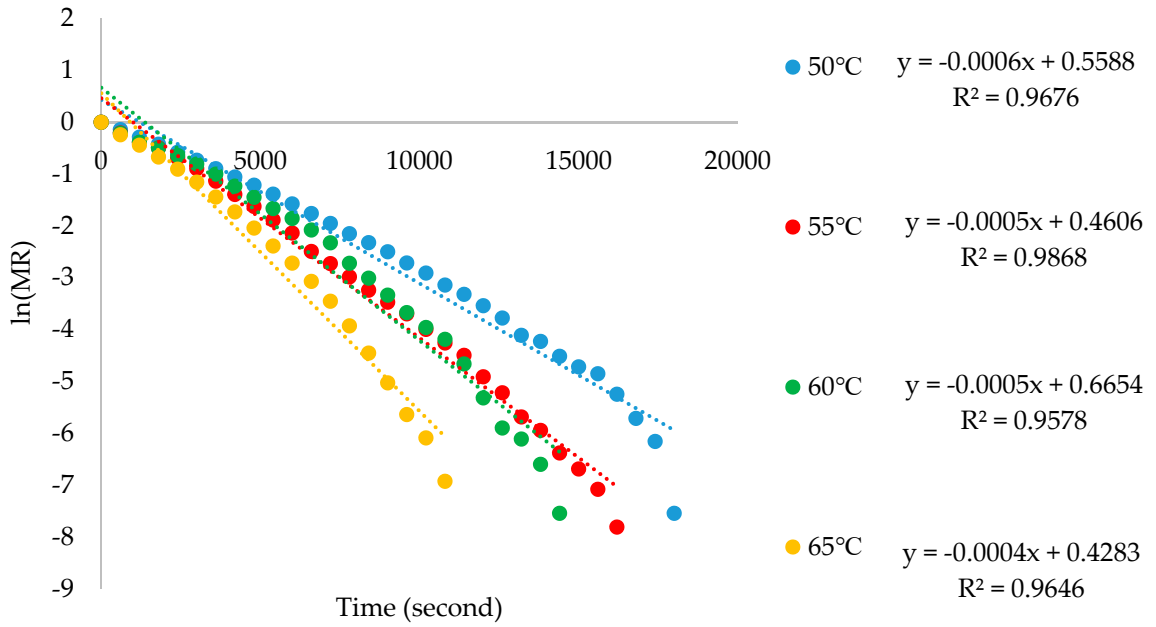


Figure 5. The change of ln(MR) by time (seconds) of sliced lemongrass.

Table 3. D_{eff} at different drying temperatures.

Temperature	Slope	D_{eff} (m/s^2)
50 °C	−0.000353784	7.64×10^{-11}
55 °C	−0.000475707	1.03×10^{-10}
60 °C	−0.000541341	1.17×10^{-10}
65 °C	−0.000684259	1.48×10^{-10}

As shown in Table 3, the D_{eff} value ranged from 7.64089×10^{-11} m^2/s to 1.47784×10^{-10} m^2/s. This result is within the normal value range of D_{eff} in typical food drying processes, which is from 10^{-12} to 10^{-6} m^2/s [14]. At temperatures of 50 °C, 55 °C, 60 °C, and 65 °C, the D_{eff} values were 7.64089×10^{-11}, 1.02741×10^{-10}, 1.16917×10^{-10}, and 1.47784×10^{-10} m^2/s respectively. This indicates that the effective diffusivity coefficient increased proportionally with temperature. At 65 °C, moisture content of citronella peaked, since higher temperature quickens evaporation of water molecules on the surface of the lemongrass material.

3.4. Estimation of Activation Energy

The graph of the change in D_{eff} value (RT_a^{-1}) is displayed in Figure 6. Using exponential regression, the activation energy of E_a of sliced lemongrass was determined to be 38.34 kJ/mol. This result is consistent with the normal range of 33.21 to 39.03 kJ/mol in the drying process of basil leaf [25]. To compare, for activation energy of fruits and vegetables, more than 90% of the activation energy values that were found in previous studies ranged between 14.42 and 43.26 kJ/mol, and 8% of the values were in the range 78.93 to 130.61 kJ/mol [25]. The present result of the activation energy for lemongrass moisture evaporation is relatively high. As a result, the separation of moisture from lemongrass could be difficult, suggesting that, in order to completely dry the lemongrass material, either drying duration or drying temperature should be set at high levels.

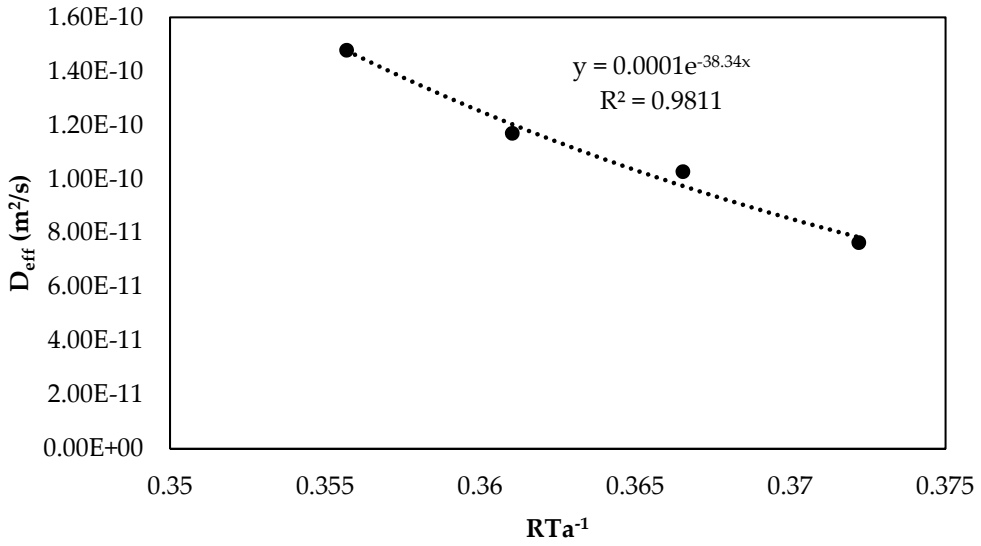


Figure 6. The relationship between change of D_{eff} and $RT_a{}^{-1}$.

4. Conclusions

Our research has identified a mathematical model suitable to describe the behavior of sliced lemongrass subjected to convection drying. In addition, the moisture diffusion coefficient and the activation energy of the material were computed. The results show that higher drying temperature is associated with shorter drying time and faster moisture removal rate. During the drying process, only initial and falling-rate periods occurred, with the absence of a constant-rate period. The Weibull model was found to be the most suitable model describing thin-layer drying by hot air.

The effective moisture diffusivity of the dried material at different temperatures was calculated and the results ranged from 7.64089×10^{-11} m^2/s to 1.47784×10^{-10} m^2/s. The moisture diffusion capability of the lemongrass material peaked at 65 °C. The calculated activation energy of lemongrass was relatively high, at 38.34 kJ/mol, indicating that more energy is needed to separate moisture from the material by drying. This may be related to the structure of the lemongrass, which consists of many layers resulting in tight links among water molecules and in turn low evaporability. These results suggest that moisture removal from the lemongrass should also take into account the remaining content of essential oils and aromatic compounds in the material.

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