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SCIENTIFIC PAPER

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THIN-LAYER DRYING MODEL OF *Cosmos caudatus*

Article Highlights

- Thin-layer mathematical models to describe the drying characteristic of *Cosmos caudatus* were studied
- Page model showed the best-fitted drying data with good statistical indicators
- D_{eff} values increased with the increase of drying temperatures (4.12×10^{-12}-24.71×10^{-12} m^2/s)
- Activation energy value calculated at 39.35 kJ/mol for drying *C. caudatus*

Abstract

Drying kinetic models and energy characteristics are well known tools to evaluate and predict the most suitable drying physiochemical conditions for a particular product. In this study, a thin-layer drying model was developed to best describe the drying kinetic behaviour of Cosmos caudatus. The drying experiments were conducted using a thermal convection oven and C. caudatus leaves were dried at five different temperatures (40, 50, 60, 70, 80 °C). Six different thin-layer drying models were proposed and applied to select the best drying model by fitting to the experimental moisture ratio data. The proposed drying models included Page, Modified Page, Lewis, Henderson-Pabis, Two Term and Weibull, and the results were statically compared and evaluated based on their goodness of fit. Among these, the Page model was found to best represent the thin-layer drying behaviour of C. caudatus with 99.76%, 5.93×10^{-5}, 9.68×10^{-5} for the coefficients determination (R^2), reduced chi-square (χ^2), and root mean square error (RMSE), respectively. The average effective moisture diffusion coefficient (D_{eff}) for the temperature 40 to 80 °C ranged from 4.12×10^{-12} to 24.71×10^{-12} m^2/s, while the activation energy (E_a) was calculated at 39.35 kJ/mol based on the Arrhenius's equation.

Keywords: thin-layer drying model, Cosmos caudatus, effective moisture diffusivity, activation energy, thermal convection oven.

Cosmos caudatus, also known as Ulam Raja (King's salad), belongs to the *Asteraceae* family. *C. caudatus* has been utilised as a medicinal plant, beginning in Latin America and afterwards developed within Southeast Asian countries such as Malaysia, Indonesia and Thailand [1,2]. In Malaysia, the plant has been cultivated as a commercial crop and widely used in many applications such as for culinary and therapeutic purposes [1]. As a local delicacy, *C. caudatus* has numerous biological properties, which have been scientifically documented, including being anti-inflammatory, antihypertensive, antidiabetic, anti-osteoporosis, antiobesity and anticancer [1-4]. The presence of important bioactive compounds (polyphenols, tannins, saponins, plant sterols, terpenes, and phenylpropanoids), minerals, and vitamins is considered as an essential quality indicator for its utilisation in many potential applications. [1,5-8].

The drying process can be considered as the most important step in downstream processing. The function of drying is to remove moisture from the fresh product, particularly from agricultural produce. Drying slows down the deterioration process by reducing contamination growth of mould or microorganisms, helps to prolong the shelf-life as well as simplifies packaging, handling, storage, and delivery [9-15]. Dif-

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ferent drying technologies have been widely introduced in the manufacturing industries. Thermal drying is one of the most conventionally used drying methods for drying biomedicinal herbal products. Compared to the traditional drying method using ambient temperature, thermal drying has greater efficiency yielding a high quality of dried product at a lower operational cost [14]. However, the thermal drying mechanism is complex, involving simultaneous heat and mass transfer for the removal of moisture content from the wet material and air [16,17]. Hence, studying the drying kinetics will aid the modelling of the drying process.

The thin-layer drying process refers to a dry layer of particles when fully exposed to drying air [16,17]. The thin-layer drying model through a mathematical solution is widely used to model drying kinetics in many agricultural products [16-21]. It is not only used to explain the drying characteristics, but also the transportation phenomena such as moisture diffusion and evaporation during the drying process [16-18]. Theoretically, thin-layer drying models are derived based on Newton's law of cooling and Fick's second law of diffusion and can be categorised into theoretical, semi-theoretical, and empirical models [16-18]. Semi-theoretical models that are modified from theoretical models have fewer assumptions and are dependent on the experimental data [16,19]. The internal factors such as drying temperature, drying air velocity, material thickness, initial moisture content, and relative humidity are the main constraints in using these models [16,20]. The Page, Newton, Midilli-Kucuk, and Henderson-Pabis models are the most used semi-theoretical models to describe the drying characteristics in many types of plant samples [16,17]. In contrast, for empirical models, the fundamental aspects are negligible, and the assumptions strongly depend on the experimental data and dimensionless analysis, meaning that the important process that takes place during drying cannot be explained [16,17,19]. Wang and Singh reported that the Weibull model is among the widely used empirical models to predict the drying process of many types of plant samples [16,17]. Among the developed thin-layer drying models, researchers found that both semi-theoretical and empirical models provided more accurate results and better prediction of drying characteristics in fruits and vegetables [17].

Although *C. caudatus* has been reported to possess numerous therapeutic potentials, the drying kinetics has yet to be studied. From an engineering point of view, the drying kinetics is crucial for the description of the drying mechanism and predicting the most suitable drying conditions. Therefore, this study has been carried out to evaluate the drying process of *C. caudatus* using a thermal convection dryer at different drying temperatures. By fitting the drying experimental data to the established mathematical drying models, the most suitable drying kinetics model of *C. caudatus* may be established. The effective moisture diffusion coefficient and activation energy of *C. caudatus* were also studied to further investigate the mass and energy characteristics throughout the drying process of *C. caudatus*.

METHODOLOGY

Preparation of *C. caudatus*

The raw materials were directly collected from the research farm of the Innovation Centre in Agritechnology for Advanced Bioprocessing (ICA), Universiti Teknologi Malaysia, Pagoh branch, Johor Bahru, Malaysia. The freshly harvested samples were randomly chosen before the drying experiment. The samples were cleaned and cut into small pieces. The initial moisture content of fresh leaf (80.5% wet basis) was measured using a moisture analyser (MX-50, A&D Instruments Ltd, Oxford shire, United Kingdom) at 105 °C for 15 min.

Preparation of drying process

The *C. caudatus* leaves were dried using a laboratory convection oven (Memmert UF110, Memmert Universal, Schwabach, Germany), and the process based on Alara *et al.* [22] with minimal modification. The oven was equipped with an adjustable temperature function that allowed the user to select the required temperature. The temperature of the ambient air was 26 °C and the relative humidity (*rh*) was in the range of 80 to 88%. The air velocity in the dryer was 1 m/s and it was measured using a digital anemometer (Digital Thermo Anemometer, Dwyer, U.S.A). Five drying temperatures of 40, 50, 60, 70 and 80 °C were used to dry the samples at a constant air velocity with three replicates for each treatment. The drying oven was firstly preheated to the chosen temperature before sample loading. An aluminium tray with a size of 22 cm×16 cm was used to place the sample (5 g) in a single layer over the tray of the dryer. Then, the sample tray was positioned in the middle of the drying chamber to ensure uniform drying. The weight loss of the sample was measured with an analytical balance (PA214C, OHAUS Corporation, USA) at selected intervals and replicated thrice. Each of the weighing processes lasted about 15 s and it was carried out near to the dryer unit. The sample was dried until a constant weight was achieved in consecutive measurements.

Determination of the thin-layer mathematical components

Moisture content

The moisture content (M) of the samples was calculated using Eq. (1) [13]:

$$M = \frac{m_t - m_{dm}}{m_{dm}} \quad (1)$$

where M, m_t and m_{dm} are the moisture content (g water/g dry matter), mass of sample (g) at a specific time and mass of dry weight (g), respectively.

Drying rate

The drying rate (DR) of the samples was calculated using Eq. (2) [13]:

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

where DR, Mt, $Mt+\Delta t$ and Δt are the drying rate (g water/g dry matter min), moisture content at time t (g water/g dry matter), moisture content at $t+\Delta t$ (g water/g dry matter), and the drying time (min), respectively.

Mathematical modeling of drying curves

The dimensionless moisture ratio (MR) was calculated from Eq. (3). Kinetic models may be used to describe the drying kinetics as a relation to the moisture transfer as shown in Table 1.

$$MR = \frac{M - M_e}{M_o - M_e} \quad (3)$$

where M is the moisture content at any drying time (dt), M_o is the initial moisture content and M_e is the moisture content at equilibrium (g water/g dry matter). For long drying, the difference of M_e is relatively small compared to M, it was therefore simplified as an Eq. (4) [13]:

$$MR = \frac{M}{M_o} \quad (4)$$

Table 1. Mathematical models for drying curves; k is drying rate constant (min^{-1}), a, b, n are constant sand t is drying time (min)

Model	Equation
Page	MR = exp ($-kt^n$)
Lewis	MR = exp ($-kt$)
Henderson-Pabis	MR = a exp ($-kt$)
Modified Page	MR = exp $(-kt)^n$
Two Term	MR = $a$exp ($-k_1t$)+$b$exp ($-k_2t$)
Weibull	MR = exp ($-[t/b]^a$)

The experimental results of MR for *C. caudatus* were fitted to the selected six thin-layer models (Table 1) and analyzed based on statistical tools such as correlation coefficient (R^2), root square error ($RMSE$) and Chi-square (χ^2), respectively. The equations are given in Eqs. (5)-(7), respectively. The higher R^2, lower $RMSE$ and χ^2 values represent a good agreement between the experimental and predicted data [13,22]:

$$R^2 = 1 - \left[\frac{\sum_{i=1}^{N} \left(MR_{\exp,i} - MR_{pre,i} \right)^2}{\sum_{i=1}^{N} \left(MR_{\exp,i} - \overline{MR}_{\exp} \right)^2} \right] \quad (5)$$

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

$$\chi^2 = \sum_{i=1}^{N} \left[\frac{MR_{\exp,i} - MR_{pre,i}}{MR_{pre,i}} \right]^2 \quad (7)$$

Estimation of the effective moisture diffusivity

The determination of the effective moisture diffusion coefficient during the drying process of *C. caudatus* was estimated using Fick's second law. The equation is expressed by Eq. (8) [22,26]:

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

For a longer drying process, $MR < 0.6$ the equation is simplified to:

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

where MR, D_{eff}, L, t and n is the dimensionless moisture ratio, the effective moisture diffusion coefficient (m^2/s), half the thickness of the initial *C. caudatus* sample (m), drying time (s) and an integer value, respectively.

Estimation of the activation energy

The activation energy is measured by the Arrhenius equation and expressed accordingly in Eq. (10) [13]:

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

D_0 is a constant in the Arrhenius equation (m^2/s), T is the temperature in degrees Kelvin (K), R is the universal gas constant (kJ/(mol K)), E_a is the activation energy (kJ/mol). By plotting D_{eff} *versus* $1/T$, the value of E_a can be determined using exponential regression.

Data analysis

The value of *M*, *MR*, *DR*, D_{eff} and E_a were calculated using Microsoft Excel software. The statistical value of R^2, *RMSE* and χ^2 of the parameter models was estimated by Excel Solver (Microsoft Office 2016).

RESULTS AND DISCUSSION

Drying characteristics of *C. caudatus*

In this study, *C. caudatus* leaves were dried at 40, 50, 60, 70 and 80 °C until equilibrium moisture content was reached. Figure 1 illustrates the drying curves in terms of moisture ratio *versus* drying time at different temperatures. While Figure 2 shows the reproducibility of the drying curve plot at 60 °C. The initial moisture content of the leaves before hitting their steady dry weight at 40, 50, 60, 70 and 80 °C was 81.11, 81.14, 82.13%, 81.30 and 82.10% (w.b), respectively. Such differences are possibly due to the different temperatures involved. As seen in Figure 1, the time to reach the equilibrium moisture content from its initial moisture content to dry *C. caudatus* leaves at 40, 50, 60, 70 and 80 °C were 100, 80, 40, 20, and 10 min, respectively. The results demonstrated that higher drying temperature (80 °C) entailed the shortest drying time. The lower the temperature (40 °C), the longer it took to dry the sample and resulted in slower drying rates. Similar observations were also reported when drying *Vernonia amygdalina* leaves and piper leaves samples [10,22]. The reproducibility of the drying curves weighted by the magnitude of standard deviations (*SD*) and the similarity between the curves. Higher reproducibility of drying curves has been tested under all temperature conditions. Figure 3 shows the example of equality between the curves.

The relation in terms of drying speeds and drying temperatures is shown in Figure 3. Significantly, the drying rate increased at an elevated drying

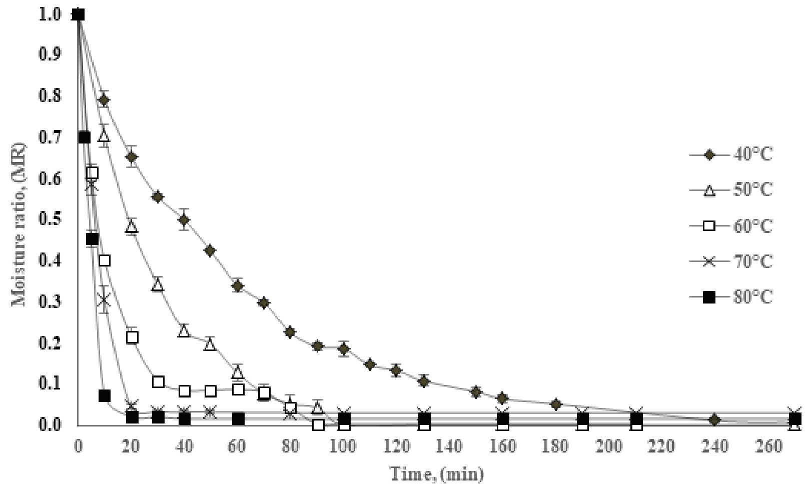

Figure 1. Drying curves of C. caudatus at different drying temperatures.

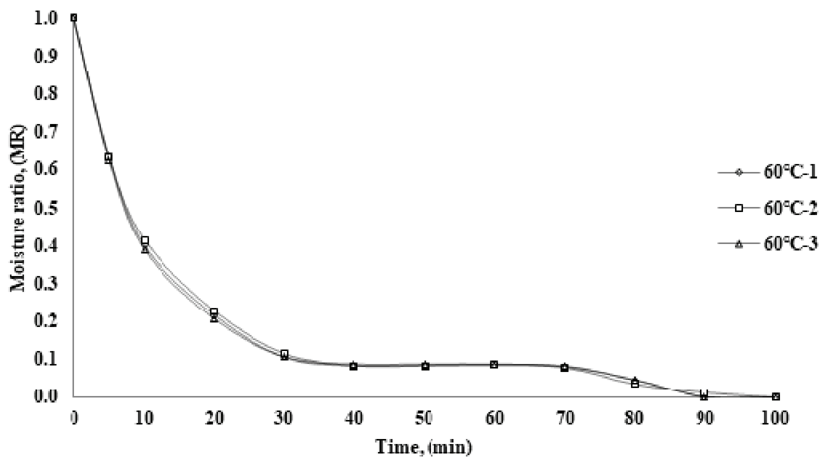

Figure 2. Example of reproducibility result (drying curve) at 60 °C.

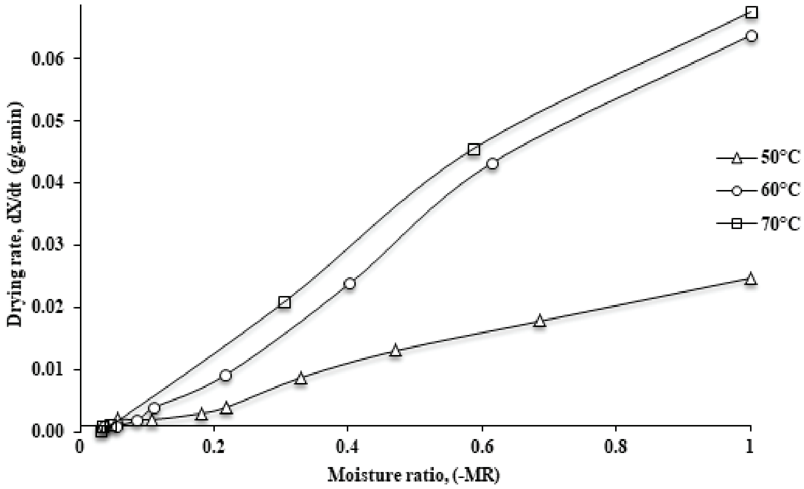

Figure 3. The drying rate curves of C. caudatus at different drying temperatures.

temperature, resulting in a rapid depletion of moisture content [15,22,26]. On the other hand, the higher drying rate accelerates further heat transfer and causes rapid removal of moisture from the material l[15,22,23]. In the drying of *Momordica charantia* slices, a fast-drying rate also occurred at 80 °C using a hot air dryer [11]. The moisture removal rate slowly reduced when approaching the equilibrium moisture content.

The entire drying process of *C. caudatus* occurred during the falling-rate period, indicating that internal moisture diffusion was the dominant factor [24]. As reported, this falling-rate period induced a shrinking effect when the surface film of the dried product appeared [17]. The results were similar to the drying behaviour reported for fruits and vegetable products [13,22,27]. As seen from Figure 3, no initial constant-rate period was observed as the evaporation process took place rapidly (negligible) at the surface of the *C. caudatus* leaves. This absence is due to the unbound moisture being insignificant in the leaves [13,14,22]. As drying continued, the material entered a second falling-rate period and then encounter a slower reduction of moisture until attaining equilibrium moisture content. The drying of *C. caudatus* stopped at this stage when no further moisture was transferred from the leaf into the surrounding air [14,15,19,24].

Thin-layer drying model

Six thin-layer models, including Page, Lewis, Henderson-Pabis, Modified Page, Two Term and Weibull, have been proposed and compared (Table 2). The suitability of the models was assessed using statistical analysis and this is shown in Table 2. The correlation coefficient (R^2) of greater than 0.9900, with the lowest root mean square error (*RMSE*), and the lowest chi-square (χ^2) values presented the most appropriate model for drying *C. caudatus.* From the results, the Page, Modified Page and Lewis models provided the appropriate results to fit the experimental data. The R^2 values of these models were estimated between 0.9900 and 0.9976. Based on the results shown in Table 1, the Page model gave an accurate prediction with the closest fit of R^2. The highest R^2 was 0.9976, with an *RMSE* of 9.68×10^{-5} and χ^2 of 5.93×10^{-5} at a drying temperature of 40 °C. Figure 4 shows the plot of the experimental data and the predicted moisture ratio by the best model for drying *C. caudatus*. The present results are in line with previous studies to best predict the drying kinetic in agricultural products such as [23] *Momordica charantia* [11], banana [29,30] and green bean [24]. This increased from 0.00246 to 0.2000 when the drying temperature was increased from 40 to 80 °C. The *k* value was linearly dependent on temperature and was consistent with the previous studies using this model [31].

The effective moisture diffusivity (D_{eff}) was calculated from the plots of ln *MR* against drying time (s) at different temperatures, and the slope of each linear regression plot was applied to estimate the D_{eff} coefficient. The change in D_{eff} coefficient at different temperatures (40 to 80 °C) is presented in Table 3. The D_{eff} coefficient showed a significant increase with an increase in temperature from 4.12×10^{-12} to 24.716×10^{-12} m^2/s. The result of this study are consistent with the reported studies, which lay within the general range of 10^{-9} to 10^{-12} m/s for food materials [13,22,32,33]. Different drying methods have been studied to investigate the change in the effective moisture diffusivity process for drying of different products. However, different drying methods, either using a thermal convection oven or a hot air dryer, also indicated an increase

Table 2. Values of the mathematical coefficients of different models for all drying temperatures

Temperature (°C)	Model	Coefficients	Statistical estimated results		
			R^2	*RMSE*	χ^2
40	Page	k = 0.0246 n = 0.9203	0.9976	9.6800×10^{-5}	5.9261×10^{-5}
50		k = 0.0542 n = 0.8648	0.9931	3.4273×10^{-4}	0.0021
60		k = 0.1529 n = 0.7402	0.9906	2.8484×10^{-4}	0.0034
70		k = 0.1300 n = 0.9001	0.9900	6.5370×10^{-4}	0.0046
80		k = 0.200 n = 0.8000	0.9904	7.6275×10^{-4}	0.0040
40	Modified Page	k = 0.1566 n = 0.1100	0.9949	2.0200×10^{-4}	0.0013
50		k = 0.3744 n = 0.1000	0.9963	1.8500×10^{-4}	0.0012
60		k = 0.3100 n = 0.3000	0.9680	7.6200×10^{-4}	0.0134
70		k = 0.400 n = 0.3020	0.9936	4.1677×10^{-4}	0.0068
80		k = 0.200 n = 0.8030	0.9885	9.1800×10^{-4}	0.0029
40	Lewis	k = 0.0180	0.9952	1.8919×10^{-5}	0.0012
50		k = 0.0400	0.9890	5.4912×10^{-4}	0.0037
60		k = 0.1200	0.9318	0.0032	0.0328
70		k = 0.1200	0.9936	4.1677×10^{-4}	0.0029
80		k = 0.1850	0.9887	8.9942×10^{-4}	0.0047
40	Henderson-Pabis	k = 0.0273 a = 1.1103	0.9187	0.0521	0.0333
50		k = 1.1046 a = 0.0605	0.9000	0.0632	0.0520
60		k = 1.0660 a = 0.0600	0.9307	0.0683	0.0301
70		k = 0.1000 a = 1.0200	0.9844	0.0010	0.0066
80		k = 0.2250 a = 1.3741	0.9000	0.1463	0.0614
40	Weibull	a = 0.0020 b = 0.1549	0.9104	0.0036	0.0200
50		a = 0.1353 b = 2.5063	0.8999	0.0045	0.0418
60		a = 0.2110 b = 2.500	0.9724	0.0013	0.0500
70		a = 0.2112 b = 1.5365	0.9860	9.1183×10^{-4}	0.0069
80		a = 0.3000 b = 1.700	0.9880	9.5623×10^{-4}	0.0049
40	Two Term	a = 0.6692 k_1 = 0.0237	0.9094	0.0667	0.0397
50		b = 0.6892 k_2 = 0.0237	0.9000	0.0041	0.0232
60		a = 0.4867 k_1 = 0.3343	0.9441	0.0030	0.0281
70		b = 0.6001 k_2 = 0.0189	0.9815	0.0012	0.0077
80		a = 0.5000 k_1 = 0.3500	0.9223	0.0054	0.0238
		b = 0.54 k_2 = 0.0665			
		a = 0.5000 k_1 = 0.200			
		b = 0.54 k_2 = 0.0665			
		a = 0.5000 k_1 = 0.4500			
		b = 0.54 k_2 = 0.0665			

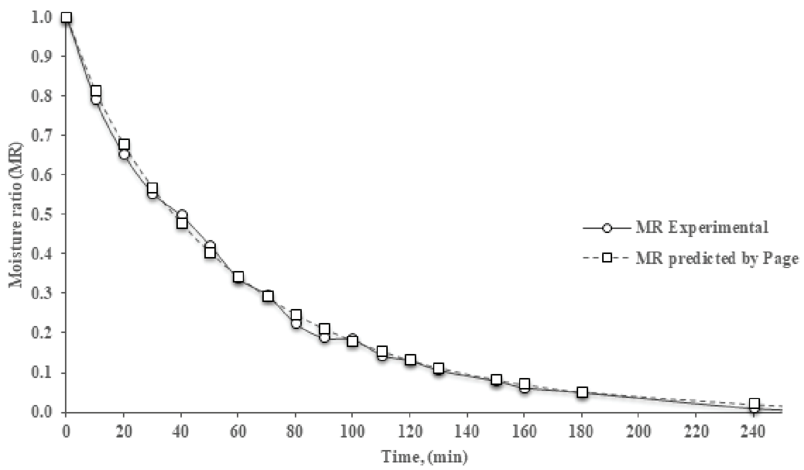

Figure 4. Experimental and theoretical moisture ratio of C. caudatus predicted by the Page model (40 °C).

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

Temperature °C	Slope (k_o)	$D_{eff}\times10^{-12}$ m^2/s	R^2
40	-0.0003	4.1194	0.9885
50	-0.0006	8.2380	0.9906
60	-0.0007	9.6110	0.9212
70	-0.0012	16.4780	0.9013
80	-0.0018	24.7160	0.9567

in the D_{eff} coefficient when the temperature was increased [13,15,22]. This is most likely because high temperature increases heat absorption in a material that increases mass transfer and drying speed [26,28]. Hence, it can be seen that the D_{eff} coefficient highly depends on the increase in the drying temperature.

Activation energy

The activation energy (E_a) represents the minimum energy required to start the removal of moisture from a material. To investigate the activation energy for drying *C. caudatus* with respect to the drying temperature, the Arrhenius equation was used to plot the exponential regression graph of ln D_{eff} against $1/T$ [13]. Based on the exponential regression plot as shown in Figure 5, the activation energy of drying *C. caudatus* was determined at 39.35 kJ/mol by thermal convection oven dryer. According to the literature review, more than 90% of the activation energy to dry a food product falls within the range between 14.42 and 43.26 kJ/mol [13,15]. The activation energy obtained in this study is also comparable to previous studies on food products such as lemongrass (38.35 kJ/mol) [13], basil (33.21 kJ/mol) [23], quinces (38.29 kJ/mol) [27], kiwifruits (34.34 to 38.07 kJ/mol) [30] and *Andrographis paniculata* (33.4 kJ/mol) [24]. Nonetheless, the difference may be due to the drying methods, materials and operating conditions during the drying process [13,21].

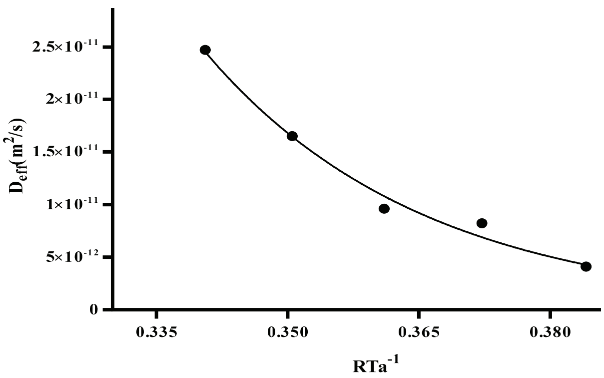

Figure 5. The relationship between change of D_{eff} and RTa^{-1} based on the Arrhenius's model (E_a =39.35 kJ/mol); $y = 2E\text{-}05e^{-39.35x}$, $R^2 = 0.9756$.

CONCLUSION

The semi-theoretical Page model was shown to be the best-fitting model for predicting the drying kinetic behavior of *C. caudatus* using a thermal convection oven. The effective moisture diffusion coefficients of the dried *C. caudatus* increased with increasing of drying temperature from 4.12×10^{-12} to 24.72×10^{-12} m^2/s and the activation energy was calculated at 39.35 kJ/mol. This study highly recommends further investigation be conducted into the physiochemical properties of *C. caudatus* and the optimisation process of drying *C. caudatus* using the selected thin-layer drying model as a benchmark. This is to improve performance and quality control of drying *C. caudatus* in terms of cost, energy and time.

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NAUČNI RAD

MODEL SUŠENJA LIŠĆA *Cosmos caudatus* U TANKOM SLOJU

Kinetički modeli sušenja i energetske karakteristike dobro su poznati kao alati za procenu i predviđanje najprikladnijih fizičkohemijskih uslova sušenja za određeni proizvod. U ovom radu, razvijen je model sušenja u tankom sloju koji najbolje opisuje kinetičko ponašanje sušenja Cosmos caudatus. Eksperimenti sušenja su izvedeni pomoću termokonvekcione peći, a lišće C. caudatus sušeni je na pet različitih temperatura (40, 50, 60, 70, 80 °C). Šest različitih modela sušenja u tankom sloju (Pejdžov, modifikovani Pejdžov, Levisov, Henderson-Pabisov, dvočlani i Vajbulov) su analizirani radi izbor najboljeg modela sušenja u odnosu na slaganje sa eksperimentalnim podacima o vlagi. Utvrđeno da je Pejdžov model najbolje opisuje kinetiku sušenja lišća C. caudatus u tankom sloju sa koeficijentom detreminacije 99,76%, redukovanim hi-kvadratom 5,93×10^{-5} *i korenom srednje kvadratne greške 9,68×10*$^{-5}$*. Prosečni efektivni koeficijent difuzije vlage za temperaturu od 40 do 80 °C kretao se od 4,12×10*$^{-12}$ *do 24,71×10*$^{-12}$ *m*^{2}*/s, dok je vrednost energije aktivacije bila na 39,35 kJ/mol na osnovu Arenijusove jednačine.*

Ključne reči: model sušenja u tankom sloju, Cosmos caudatus, efektivna difuznost vlage, energija aktivacije, termokonvekciona peć.