

RESEARCH ARTICLE

Drying kinetics characteristics and model fitting of exocarpium citreic grandis microwave vacuum drying

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Exocarpium citreic grandis is the immature outer peel of grapefruit and contains active ingredients such as naringin and rhoifolin. The drying temperature can have a significant effect on the constituents. In order to explore the rules of moisture and quality changes in exocarpium citreic grandis during microwave vacuum drying, this study investigated the effects of different microwave intensities (0.75, 1.25, 1.50, 1.75, 2.00 W/g) on the drying characteristics and main component content of exocarpium citreic grandis. The results showed that, at a microwave intensity of 0.75 W/g, the drying process was controlled by a constant speed stage. At microwave intensities of 1.25 W/g and 1.50 W/g, the drying process was divided into three parts including acceleration stage, constant speed stage, and deceleration stage. When the microwave intensity increased to 1.75 W/g and 2.00 W/g, the drying process was divided into acceleration and deceleration stages with no constant speed drying stage. The resulted data suggested that the thin-layer drying kinetics model of exocarpium citreic grandis was fitted. The selection principle of the model was analyzed from four aspects including modification of the original model, transformation of dependent and independent variables, complex model, and initial and terminal conditions. Lewis, Page, Two-term exponential, and Weibull distribution models were selected for fitting analysis. The optimal thin-layer drying models were the Page model and the Weibull distribution model. The scale parameter α of the Weibull distribution function decreased with the increase in microwave intensity. The shape parameter β underwent slight changes with moisture migration mechanisms being the combined effect of surface moisture evaporation and internal moisture diffusion. The effective moisture diffusion coefficient D_{eff} was increasing with the increase in microwave intensity. The activation energy Ea for microwave vacuum drying of exocarpium citreic grandis was 1.6799 kJ/mol·k. Microwave power had a significant effect on flavonoid compounds. When the microwave power was low, the main component content decreased with the increase in microwave intensity, but when the microwave power increased to 2.00 W/g, the drying temperature would produce small molecular phenolic compounds, leading to an increasing trend in total flavonoids, naringin, and rhoifolin contents. With the increase in microwave intensity, the surface of exocarpium citreic grandis presented more porous honeycomb structures, and the larger the pore size, the more evident the puffing effect. The study confirmed that microwave vacuum drying technology was suitable for the production and processing of exocarpium citreic grandis, which provided technical support for the high efficiency and high quality industrial production.

Keywords: microwave vacuum drying; exocarpium *Citri Grandis*; Weibull distribution function; kinetic model; effective moisture diffusion coefficient.

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Introduction

Exocarpium citreic grandis has a long medicinal history and is a native medicinal material in the Lingnan region (Guangdong and Guangxi provinces of China). *Exocarpium citreic grandis*, belonging to the *Rutaceae* family, is the dried outer layer of immature or nearly ripe citrus fruit. It has been attributed with functions such as dispersing cold and dampness, regulating Qi (a concept originating from traditional Chinese culture that refers to a vital energy or life force that flows through all living things) and resolving diseases, dissolving phlegm, strengthening the spleen, aiding digestion, and relieving pain and fever [1, 2]. According to the fingerprinting results of flavonoid components, naringin and rhoifolin are the main constituents of *exocarpium citreic grandis*, accounting for over 80% of the content. With a high moisture content of approximately 80%, fresh *exocarpium citreic grandis* is challenging to store and prone to serious loss of active ingredients. Drying is one of the most common preservation methods. Many scholars have investigated the medicinal value of the *exocarpium citreic grandis*. Naringin, the primary component of *exocarpium citreic grandis*, exhibits various biological activities such as antioxidation, blood sugar reduction, blood lipid reduction, anti-atherosclerosis, and anti-tumor [3]. Liu *et al.* conducted hot air drying on *exocarpium citreic grandis* in the temperature range of 50–70°C and found that the effective diffusion coefficient of moisture in *exocarpium citreic grandis* ranged from 0.2311×10^{-7} to 0.7865×10^{-7} m²/s, positively correlating with drying temperature and slice thickness [4]. Research related to the kinetics and quality of microwave vacuum drying of *exocarpium citreic grandis* has not been found and requires further exploration.

Microwave vacuum drying is a new generation processing method that removes moisture from materials under vacuum conditions with the aid of microwave heating. It combines the advantages of instant microwave heating and reduced boiling points of water in a vacuum

environment. This drying technique has been applied to the processing of agricultural products like beef [5], *Angelica sinensis* [6], garlic [7], lemon [8], potato [9], lotus seed [10], *etc.* The quality and energy consumption of the dried products are closely related to the internal moisture diffusion mechanism, but observing moisture changes during the drying process in real-time is challenging. Some scholars have proposed empirical and semi-empirical mathematical models, such as Page and Lewis models, to describe the moisture variation during drying. Although these models can simulate moisture variation well, the physical meaning of model parameters remains unclear, hindering the understanding of heat and mass transfer during drying and the selection of drying conditions [11]. In contrast, the scale parameter (α) and shape parameter (β) in the Weibull distribution function relate to the heat and mass transfer in the drying process, and when combined with drying methods and conditions, they can be used to estimate the moisture diffusion coefficient and determine whether there is an acceleration stage [12]. Scholars have utilized the Weibull distribution function to model Sichuan pepper [13], Poria [14], potato [15], lotus pollen [16], *etc.*, achieving good results in predicting, controlling, and optimizing the drying process. However, the application of the Weibull distribution function to the *exocarpium citreic grandis*' drying kinetics has not yet been explored.

This study took *exocarpium citreic grandis* as the research object and selected appropriate thin-layer drying models for the experimental data to study the impact of microwave intensity on the drying characteristics of *exocarpium citreic grandis*. Based on the Weibull distribution function, the microwave vacuum drying process of *exocarpium citreic grandis* was simulated, the effective moisture diffusion coefficient and activation energy in its drying process were calculated, and the effects of microwave intensity on the main constituents including total flavonoids, naringin, and rhoifolin were analyzed. Furthermore, the internal microstructure of

exocarpium citreic grandis was analyzed to explore the mechanism of rapid microwave drying, providing theoretical support for predicting and controlling the drying process of the exocarpium citreic grandis. This study focused on drying efficiency and high quality, which used the new drying technology of microwave vacuum to carry out drying tests. Therefore, the study provided scientific support for the application of microwave technology in the field of traditional Chinese medicine.

Materials and Methods

Sample preparation and treatment

Exocarpium citreic grandis was harvested from Luchuan exocarpium citreic grandis manor (Luchuan, Guangxi, China) and selected with consistent maturity, uniform size, and subsequently stored in a refrigerator. The exocarpium citreic grandis slices were dried in an oven at 105°C until no further changes in mass were observed, resulting in a moisture content of 78% on a wet basis. According to the pharmacopoeia, exocarpium citreic grandis dried to a moisture content of less than 11% on a wet basis was considered a qualified product. A total of 2,000 g of exocarpium citreic grandis slices were evenly spread across 6 rotating baskets inside the WZD6 Microwave Vacuum Drying Sterilization Furnace (Nanjing Sanle Microwave Technology Development Co. Ltd., Nanjing, Jiangsu, China) with a slice thickness of 6 mm. The drying experiment was conducted at different microwave intensities of 0.75 W/g, 1.25 W/g, 1.50 W/g, 1.75 W/g, 2.00 W/g, and a relative vacuum degree of -90 kPa. The furnace's top was equipped with a temperature collection device to monitor the surface temperature of the rotating exocarpium citreic grandis, displaying and storing the data in real-time. Samples were weighed every 10 minutes during the experiment. Due to the rapid temperature increase caused by microwaving, the effect of weighing on the material temperature was negligible, and each test was repeated three times, taking the average value.

Drying parameter calculation

The moisture ratio (MR) represented the change in water content during drying and could be simplified as:

$$MR = M_t / M_0 \quad (1)$$

where, M_t was the moisture content of exocarpium citreic grandis at time t on a dry basis (g/g). M_0 was the initial moisture content of exocarpium citreic grandis on a dry basis (g/g). The drying rate D_R (kg/kg·min) was a parameter measuring the extent of exocarpium citreic grandis moisture variation over time and was calculated as follows:

$$D_R = -(m_{(t+\Delta t)} - m_t) / \Delta t \quad (2)$$

where, $m_{(t+\Delta t)}$ and m_t were the moisture contents on a dry basis at times $t+\Delta t$ and t , respectively (kg/kg).

Weibull distribution model

The Weibull distribution model was a mathematical model obtained using the Weibull distribution in statistics, given by equation (3).

$$MR = \exp \left[- \left(\frac{t}{\alpha} \right)^\beta \right] \quad (3)$$

where α was the scale parameter, roughly corresponding to the time required for the material to lose 63% of its moisture (min). β was the shape parameter, related to the initial drying rate and the mechanism of moisture migration within the material. t was the drying time (min) [11].

Effective moisture diffusion coefficient

The drying characteristics were described by using Fick's diffusion equation with the simplified computation expressed as:

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

where, D_{eff} was effective moisture diffusion coefficient (m^2/s). L was sample thickness (m). A linear fitting of $\ln MR$ was used to determine the effective moisture diffusion coefficient. When Fick's second law was applicable during the falling-rate drying phase, its application became highly limited in many drying processes that included increasing, constant, and falling-rate stages. Using the Weibull distribution function ignored the characteristics of moisture migration, regardless of which phase the drying process was in, as calculated by the following:

$$D_{cal} = \frac{r^2}{\alpha} \quad (5)$$

where, r was effective radius of longan (13 mm).

$$D_{eff} = \frac{D_{cal}}{R_g} \quad (6)$$

where, R_g was constant related to geometric dimensions [17].

Calculation of drying activation energy

The drying activation energy represented the microwave energy required to remove a mole of moisture during the drying process. A higher value indicated more energy was needed. The relationship between the internal moisture diffusion in exocarpium citreic grandis and the microwave power followed the Arrhenius equation as calculated by:

$$D_{eff} = D_0 \cdot \exp\left(-\frac{E_a \cdot m}{p}\right) \quad (7)$$

The organized estimation formula for the moisture diffusion coefficient was as follows:

$$D_{cal} = R_g \cdot D_0 \cdot \exp(E_a \cdot m/p) \quad (8)$$

where, D_0 Arrhenius was pre-exponential factor, a constant (m^2/s). E_a was material drying activation energy (kJ/mol). m was material mass (g). P was microwave power (W) [18].

Selection of the thin-layer drying model

The thin-layer drying mathematical models were abundant with as many as 99 variations available for use [19]. This study aimed to select an appropriate mathematical model suitable for exocarpium citreic grandis drying, based on the following criteria:

(1) Model modification

Modifying thin-layer drying models helped reduce the interrelationship of parameters, making the model more easily convergent, while maintaining the same fitting characteristics as the original model. The Page model was analyzed and modified as below.

$$MR = \exp(-kt^n) \quad (9)$$

$$MR = \exp[-(Kt)^n] \quad (10)$$

where, MR represented the moisture ratio, the dependent variable, and t denoted the drying time, the independent variable. There were two parameters in the model, k and n , which determined the trend of the drying curve. Equation (10) referred the modified Page model and its relationship among parameters $k = K^n$. Under the condition of the same value of n , the two models had the same degree of fit, meaning both models exhibited identical coefficients of determination and mean square errors. By fitting the two models to experimental data at 0.75 W/g, the values for both models were found to be 0.00169, the R^2 values were 0.0011, and the χ^2 values were 1.73624, with only the k value differing between the two models. Thus, only one of the original or modified models needs to be selected for fitting.

(2) Transformation of dependent or independent variables

Some models describing drying data required the transformation of independent and dependent variables, followed by a fitting analysis. Taking the Dincer model as an example, the model transforms the MR function into $\ln MR$. The model of Dincer was shown in equation (11):

$$MR = G \cdot \exp(-st) \quad (11)$$

where G and s were the model parameters. Using the test data of 0.75 W/g for error analysis of this model, although the transformed model was successfully fitted, the R^2 value was only 0.85248. The alteration of the original experimental data by the model and the subsequent transformation of the dependent variable into heteroscedasticity led to a failure in model prediction. Although the fitting was successful, the model lost significance.

(3) Complex models

Complex models referred to those with more than three parameters. In this research, the modified Henderson and Pabis model was exemplified as shown in equation (12).

$$MR = a \exp(-kt) + b \exp(-gt) + c \exp(-ht) \quad (12)$$

The formula consisted of six parameters including a , b , c , k , g , and h . Fitting this model to the experimental data of 0.75 W/g led to an erroneous association between the model parameters and the probability P value of the parameter values, all greater than 0.05, resulting in a fitting failure, which suggested that an excessive number of parameters could lead to unreliable fitting values. Similarly, the fitting of the Two-term exponential model to the experimental data of 0.75 W/g resulted in P values for the two parameters a and k of less than 0.0001, and an R^2 value of 0.97874, indicating reliable model parameter fitting. Therefore, in selecting a drying model, priority should be given to simple models with three or fewer parameters.

(4) Initial and terminal conditions

The drying model should satisfy both the initial and terminal conditions, namely, when $t = 0$, $MR = 1$, and when $t \rightarrow \infty$, $MR \rightarrow 0$. Models that did not meet these initial and terminal conditions should not be considered. For instance, consider the Logarithmic model, represented by equation (13):

$$MR = a \cdot \exp(-k \cdot t) + c \quad (13)$$

where a and c were the model parameters, when $t = 0$, $MR = a + c = 1$, and upon fitting the experimental data of 0.75 W/g, the parameter values were $a = 3.17285$, $k = 0.00565$, and $c = -2.15089$. As the sum of $a + c$ did not equal to 1, despite an R^2 value of 0.99296, indicating a high degree of model fit, the initial conditions were not met, thus rendering the model devoid of practical physical meaning. Following a comprehensive analysis, this study selected four thin-layer drying models including Lewis, Page, Two-term exponential, and the Weibull distribution function.

Determination of naringin and rhoifolin content

Naringin standard (91.7%) and rhoifolin (95.5%) were purchased from the China Food and Drug Inspection and Testing Center (Beijing, China). *Exocarpium citreic grandis* was finely grounded and sifted through a No. 3 sieve (pore diameter: $355 \pm 13 \mu\text{m}$). 0.2 g of powder was weighed precisely and placed in a 50 mL centrifuge tube. After 20 mL of methanol was accurately added, the mixture was treated with ultrasound by using KQ-250DB CNC Ultrasonic Cleaner (Kunshan Ultrasonic Instruments Co. Ltd., Kunshan, Jiangsu, China) for 30 minutes. After cooling and reweighing, methanol was used to make up the lost weight. The mixture was shaken, filtered through a 0.45 μm microporous filter membrane, and the filtrate was used as the sample solution. Naringin (22.42 mg) and rhoifolin (9.73 mg) standards were precisely weighed and placed in 10 mL volumetric flasks before dissolved in methanol to 50 mL to prepare a mixed standard curve. Agilent 1260 high-performance liquid chromatograph (Agilent Technologies, Santa Clara, CA, USA) with a DAD detector and quaternary pump gradient elution was applied. Separation was achieved through a CAPCELL PAK C18 MG II (4.5 mm \times 250 mm, 5 μm) chromatography column with the conditions as 25°C column temperature and 330 nm wavelength. The mobile phase used methanol-glacial acetic acid aqueous solution system with methanol as phase A and glacial acetic acid

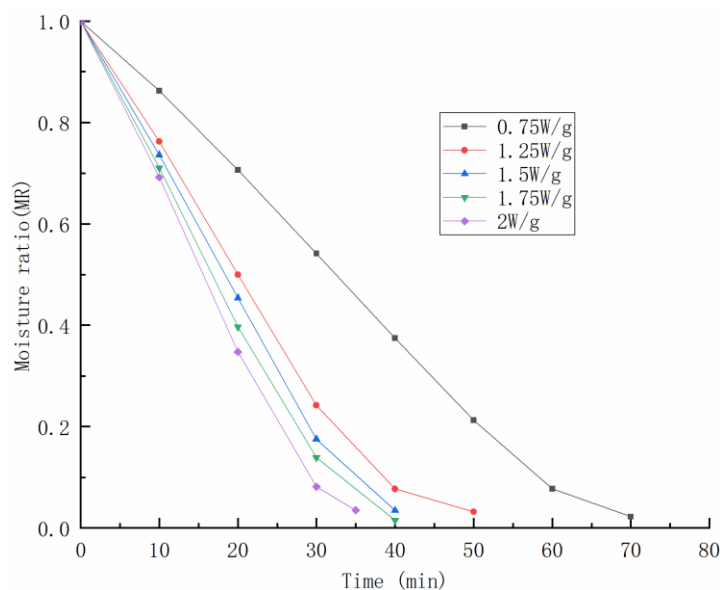


Figure 1. The drying curve of exocarpium citreic grandis slices at different microwave intensities.

aqueous solution as phase B (4 → 61), gradient elution (0 to 18 min, 25% A to 47% A) at a flow rate of 1.00 mL/min. The injection volume was 10 μ L. By using peak area as the x-axis and concentration as the y-axis, linear regression analysis was conducted. The regression equation for naringin was $y = 2,037.9x - 393.11$, $r = 0.9996$, with a linear range of 0.2570 to 1.2849 mg/mL. For rhoifolin, the regression equation was $y = 1,534.7x - 132.07$, $r = 0.9997$, with a linear range of 0.0232 to 0.1162 mg/mL, indicating a good linear relationship [20].

Statistical analysis

The experimental data were subjected to regression and variance analysis utilizing Origin 8.0 software (OriginLab, Northampton, MA, USA). The goodness of fit was assessed by determination coefficient (R^2), chi-square value (χ^2), and root-mean-square error (RMSE), where a higher R^2 and lower χ^2 and RMSE indicated a more robust fit of the model [21].

Results and discussion

Analysis of drying characteristics of exocarpium citreic grandis under microwave vacuum drying

The drying curve for exocarpium citreic grandis slices with a thickness of 6 mm and a vacuum degree of -90 kPa was illustrated in Figure 1. At microwave intensities of 0.75, 1.25, 1.50, 1.75, and 2.00 W/g, the drying time was significantly reduced with the drying time at 2 W/g being reduced by 50% compared to that at 0.75 W/g. A higher microwave intensity led to an increased absorption of microwave energy by the polar water molecules in the exocarpium citreic grandis per unit time, accelerating the vaporization rate, and consequently shortening the drying time. From the drying rate curve shown in Figure 2, the drying rate of exocarpium citreic grandis exhibited substantial changes with increasing microwave intensity. At a lower microwave intensity of 0.75 W/g, the drying rate reached 4.85 g/g·min after 10 minutes followed by a constant drying phase lasting for 50 minutes. The drying rate slowly fluctuated within the range of 5.54 to 5.93 g/g·min, and the deceleration drying phase lasted for about 10 minutes. When the microwave intensity increased to 1.25 and 1.50 W/g, the drying process was divided into three stages as acceleration, constant drying, and deceleration. After 10 minutes of acceleration, both entered a constant drying stage of about 20 minutes with

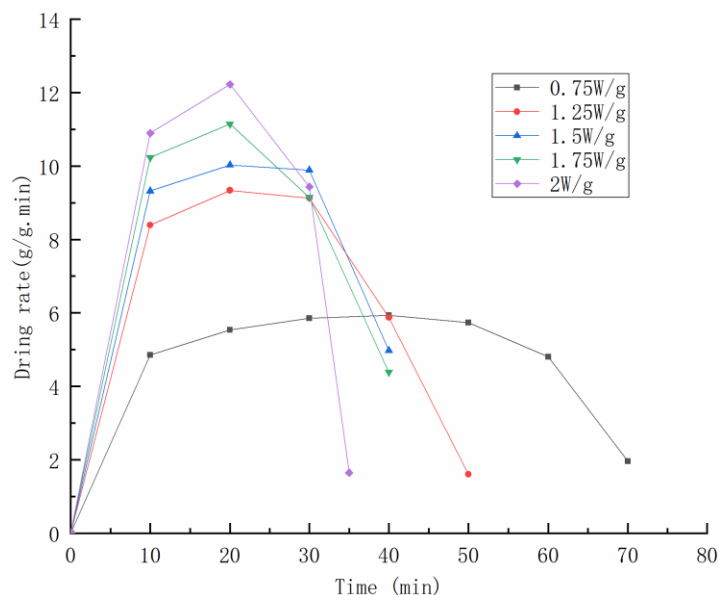


Figure 2. Variation of drying rate of exocarpium citreic grandis slices at different microwave power levels.

maximum drying rates of 9.34 g/g·min and 10.02 g/g·min, respectively. At the microwave intensities of 1.75 W/g and 2.00 W/g, the drying process was divided into acceleration and deceleration stages without a constant drying phase. Both reached maximum drying rates of 11.15 g/g·min and 12.23 g/g·min, respectively in the first 15 minutes followed by a deceleration phase. It was hypothesized that, when the microwave intensity was lower, the exocarpium citreic grandis absorbed less microwave energy, which resulted in smaller pressure and temperature differences induced by water evaporation, and the internal moisture was expelled by diffusion, leading to a slow rise in drying rate. Upon entering the constant drying phase, the microwave energy absorption and heat energy consumed by water evaporation were balanced, resulting in minor changes in drying rate and prolonged drying time. With increased microwave intensity, the absorbed microwave energy in the exocarpium citreic grandis increased, the water evaporation rate accelerated, and the vapor pressure difference and temperature difference grew, causing not only diffusion of internal moisture but also internal evaporation and liquid water in three ways of discharge, significantly increasing the

drying rate and shortening the overall drying process.

Analysis of thin-layer drying models for exocarpium citreic grandis

By using the experimental data, fitting analysis was conducted for the models (Table 1). The parameters for the four thin-layer drying models of Lewis, Page, Two-term exponential, and Weibull distribution were obtained with respective average coefficients of determination R^2 values of 0.9427, 0.9949, 0.9891, and 0.9949. The average chi-squared (χ^2) values were 0.008792, 0.000768, 0.00203, and 0.000768, respectively, and the average root mean square error (RMSE) values were 0.08489, 0.02076, 0.03519, and 0.01996, respectively. It was found that, within the experimental data range, the Page model and Weibull distribution exhibited nearly identical fitting characteristics with optimal fitting effects, differing only in the parameter values of the two models. Transformations applied to the Weibull distribution model resulted in an equation as:

$$MR = \exp \left[- \left(1/\alpha \right)^\beta \times t^\beta \right]$$

Table 1. Model fitting results.

Model Name	Microwave Intensity (W/g)	Parameters	R ²	RMSE	χ ²
Lewis	0.75	k=0.02651	0.9259	0.08499	0.00979
	1.25	k=0.04342	0.9512	0.08602	0.0074
	1.50	k=0.04717	0.9443	0.08359	0.00874
	1.75	k=0.05261	0.9461	0.08428	0.00888
	2.00	k=0.0579	0.9459	0.08557	0.00915
Page	0.75	k=0.00178, n=1.73624	0.9917	0.01354	0.0011
	1.25	k=0.00554, n=1.63539	0.9971	0.01866	4.3536E-4
	1.50	k=0.0056, n=1.68249	0.9945	0.02555	8.7036E-4
	1.75	k=0.0069, n=1.68642	0.9946	0.02589	8.9357E-4
	2.00	k=0.0064, n=1.73070	0.9968	0.02016	5.4211E-4
Two-term exponential	0.75	a=2.082160, k=0.0439	0.9784	0.02728	0.00281
	1.25	a=2.09784, k=0.07089	0.9911	0.03293	0.00136
	1.50	a=2.12577, k=0.07888	0.9986	0.04046	0.00218
	1.75	a=2.16345, k=0.08775	0.9869	0.04012	0.00215
	2.00	a=2.24205, k=0.09815	0.9903	0.03514	0.00165
Weibull distribution	0.75	α=38.2969, β=1.73625	0.9917	0.01343	0.0011
	1.25	α=23.9806, β=1.63539	0.9971	0.01866	4.3536E-4
	1.50	α=21.79032, β=1.68248	0.9945	0.02555	8.7036E-4
	1.75	α=19.99806, β=1.68643	0.9946	0.02198	8.9357E-4
	2.00	α=18.51349, β=1.7307	0.9968	0.02016	5.4211E-4

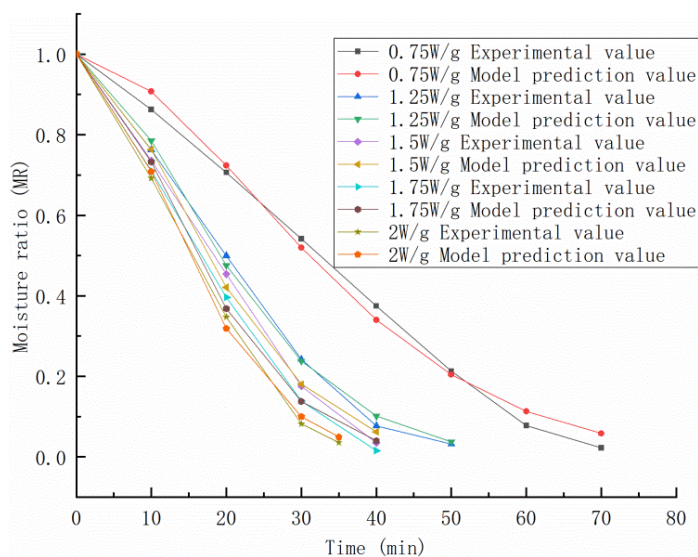


Figure 3. Drying curve comparison between experimental values and Weibull model predictions at different microwave intensities.

The parameter $-(1/\alpha)^\beta$ in the model corresponded to -k in the Page model, and β corresponded to n. Following the transformation, the two models exhibited identical structural compositions, resulting in completely similar

fitting characteristics. In the Weibull distribution model, each parameter carried a specific physical meaning. The fitting curve of the Weibull distribution function was illustrated in Figure 3. The model aligned well with the experimental

Table 2. Effective moisture diffusion coefficients.

Microwave Intensity (W/g)	D_{eff} ($\times 10^{-6}$ m ² /s)	D_{cal} ($\times 10^{-6}$ m ² /s)	R_g
0.75	0.7418	1.7553	1.3021
1.25	1.0381	2.8002	2.8002
1.5	1.1897	3.0852	2.5933
1.75	1.4046	3.3617	2.3934
2	1.4582	3.6194	2.4821

results, providing an accurate prediction and simulation for the drying experiment.

Analysis of Weibull distribution model for drying

(1) Parameter analysis

The scale parameter α , representing the time required to complete 63% of the drying process, was associated with the drying rate. As indicated in Table 1, the scale parameter α decreased by 51.7% from 38.2969 minutes to 18.5135 minutes as the microwave intensity increased from 0.75 W/g to 2.00 W/g. As the microwave intensity increased, the internal heat and mass transfer of the exocarpium citreic grandis substance intensified, the drying rate increased, and the microwave intensity had a significant impact on the scale parameter. The shape parameter β , key to explaining the moisture migration mechanism, signified the dominating role of internal moisture diffusion when its value ranged from 0.3 to 1, which indicated that the deceleration stage controlled the entire drying process. When the shape parameter β was greater than 1, both surface moisture evaporation and internal moisture diffusion jointly contributed to the moisture migration mechanism. In this study, the value β of the shape parameter exhibited a minor variation between 1.63539 and 1.73625, reflecting the influence of microwave intensity to the β value, albeit insufficient to alter the moisture migration mechanism. An increase in microwave power led to a slight increase in the β value, suggesting the existence of a lag phenomenon during the initial drying phase.

(2) Analysis of effective moisture diffusion coefficient and activation energy

The moisture diffusion coefficient is a dynamic quantity that describes the intrinsic moisture

diffusion characteristics of the material. It is associated not only with the material's structure, composition, moisture state, but also with the drying method and conditions, involving molecular diffusion, adsorption kinetics, and capillary flow. The effective diffusion coefficient and estimated effective diffusion coefficient were shown in Table 2. Based on the estimated effective moisture diffusion coefficient D_{cal} ranging between 1.7553×10^{-6} to 3.6194×10^{-6} m²/s, and the effective moisture diffusion coefficient D_{eff} obtained according to Fick's second law, ranging from 0.7418×10^{-6} to 1.4582×10^{-6} m²/s, both increased with the intensification of microwave power. The greater the microwave intensity, the more vigorous the movement of water molecules, leading to an accelerated moisture migration rate and an increase in the effective diffusion coefficient. Numerically, D_{cal} was greater than D_{eff} . Analysis suggested that Fick's second law applied to the drying process controlled by the falling rate stage, overlooking the moisture migration caused by the constant rate and increasing rate stages. In this study, at lower microwave intensity, the drying process was dominated by the constant rate drying stage, hence, there was a significant numerical discrepancy between the two. The use of the estimated effective moisture diffusion coefficient D_{cal} did not require consideration of the specific drying stage. The geometrical parameter R_g , related to the geometric size of exocarpium citreic grandis, varied between 1.3021 and 2.4821. Analysis indicated that the microwave vacuum drying process produced an expansion effect in exocarpium citreic grandis, leading to significant changes in the internal structural organization. The activation energy represents the microwave

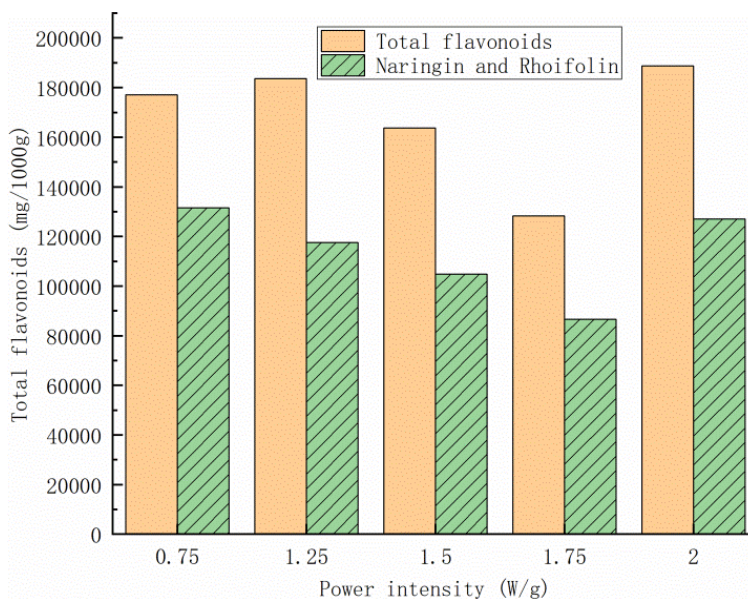


Figure 4. Impact of microwave power density on main component content.

energy required to remove one mole of water during the drying process, with a higher value indicating a greater energy requirement. By fitting equation 8, the activation energy (E_a) for microwave vacuum drying was found to be 1.6799 kJ/mol·K, which was comparable to the activation energies of other materials reported by previous study [18].

(3) Analysis of main content in exocarpium citreic grandis

The intensity of microwave radiation significantly affected the total flavonoids, naringin, and rhoifolin in exocarpium citreic grandis (Figure 4). Within the total flavonoid content of exocarpium citreic grandis, naringin and rhoifolin constituted a substantial proportion. Under microwave intensities of 0.75, 1.25, 1.50, 1.75, and 2.00 W/g, the respective proportions of naringin and rhoifolin to total flavonoids were 74.32%, 64.04%, 64.03%, 67.5%, and 67.35%, respectively. The content of these primary components was higher when the microwave intensity was relatively low, at 0.75 and 1.25 W/g. As the microwave intensity increased, the main content exhibited a decreasing trend with the total flavonoids, naringin, and rhoifolin reaching their lowest levels at 1.75 W/g. The analysis

revealed that, when the microwave intensity was low, the drying temperature stayed below 50°C, effectively preserving the active constituents of exocarpium citreic grandis. With an increase in microwave intensity, drying at the peak temperatures of 53°C for 1.5 and 1.75 W/g enhanced the flavonoid activity, accelerated the degradation rate, and resulted in a decline in flavonoid content. However, when the microwave intensity escalated to 2.00 W/g and the drying temperature reached 60°C, the total flavonoid content conspicuously increased. Within the experimental parameters of this study, the total flavonoid content exhibited a downward trend with an increase in microwave intensity. However, when the drying temperature raised to 60°C, the total flavonoid content significantly increased. This finding aligned with the conclusions in studies by Jeong *et al.* who discovered that high-temperature treatments (100°C and 150°C) led to an increase in both total phenol and flavonoid contents in orange peel [22]. Specifically, the treatment at 150°C for 30 minutes resulted in the formation of some novel small-molecule phenolic compounds. The results of this research were consistent with the findings of previous studies [22, 23].

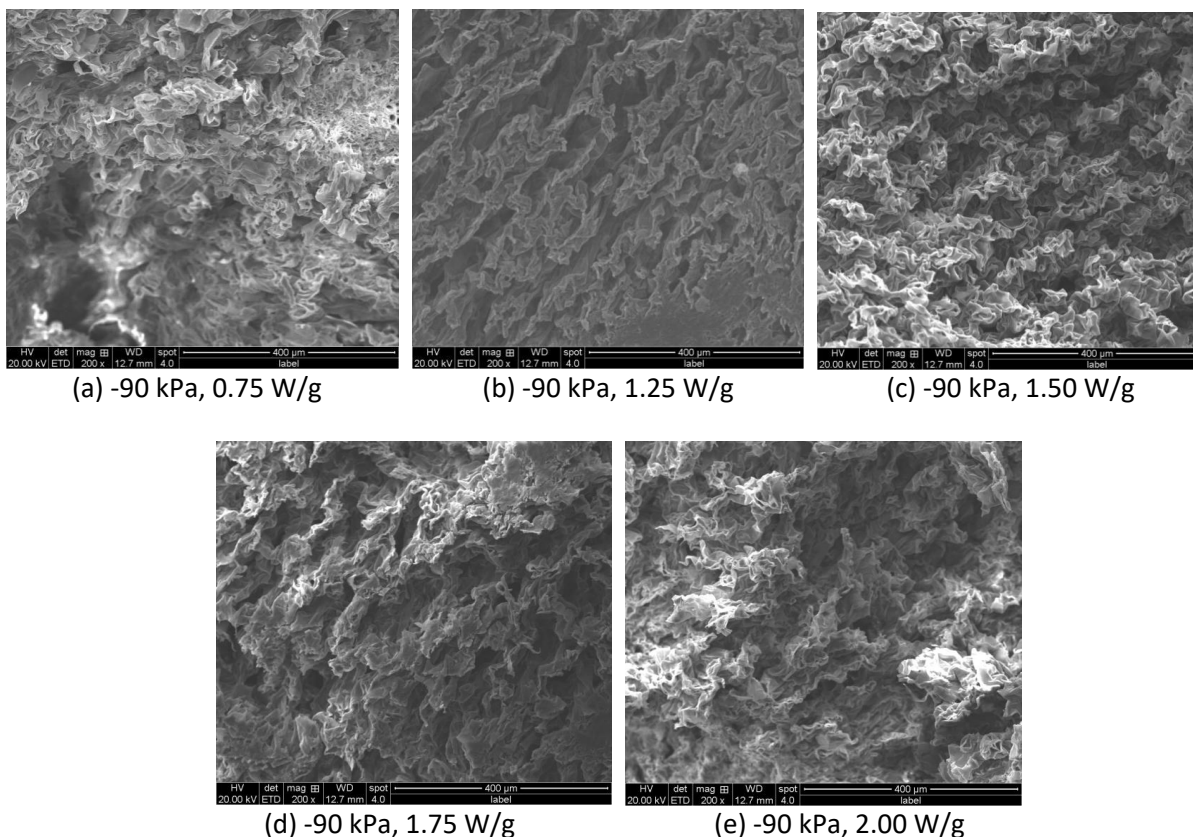


Figure 5. The Scanning Electron Microscopy (SEM) images of exocarpium citreic grandis.

Microstructure analysis

Figure 5 displayed the images of exocarpium citreic grandis obtained from Quanta 250 Scanning Electron Microscopy (FEI Company, Hillsboro, OR, USA) at magnified 200 times and different microwave intensities. The images showed that different microwave intensities led to pronounced variations in the microstructure of exocarpium citreic grandis. When the microwave intensity was relatively low at 0.75 W/g, the cellular structure was clear, and there was no local occurrence of cellular rupture or swelling phenomena. As the microwave intensity continued to escalate, a honeycomb-like structure became prominent, the cellular structure underwent destruction and deformation. The holes which were formed by the instantaneous evaporation of water vapor became increasingly large. The results were attributable to the expansion effect produced by the microwave. As microwave intensity

increased, the resistance to moisture transmission decreased, elucidating the mechanism of the increasing drying rate with microwave intensity.

Conclusion

Microwave intensity had a significant impact on the drying kinetics of exocarpium citreic grandis. The drying curve indicated that, at a microwave intensity of 0.75 W/g, the constant-rate drying stage controlled the entire drying process. When the intensity increased to 1.25 W/g and 1.50 W/g, the drying process could be divided into three parts as acceleration stage, constant-rate stage, and deceleration stage. As the intensity reached 1.75 W/g and 2.00 W/g, the drying process comprised an acceleration stage and a deceleration stage with no constant-rate drying phase. The drying rate increased with the

enhancement of microwave intensity. The optimization of model fitting yielded the optimal thin-layer drying models of the Page model and the Weibull distribution model. Both models, upon transformation, shared the same structure and, therefore, exhibited identical fitting characteristics. Both models possessed a maximum R^2 value of 0.9938 and a minimum RMSE value of 0.02339. The Weibull distribution model could accurately describe the drying process. The effective diffusion coefficient of exocarpium citreic grandis increased with microwave intensity. Microwave power had a pronounced effect on flavonoid compounds. When the microwave power was relatively low, the main content decreased with increasing microwave intensity. However, when the microwave power elevated to 2.00 W/g, the high temperature generated small-molecule phenolic compounds from the flavonoids, resulting in an increase in the total flavonoids, naringin, and rhoifolin contents. Microstructure observation revealed that microwave vacuum drying induced a swelling effect on the material, thereby accelerating the drying process.

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