Scientific Research

Study on Thin Layer Drying of *Rosa laevigata* Michx

Bing Lin[1,2,3], Ying Wang[1], Ying Zhou[1,2,3], Zhi Zhao[1,3]

[1]College of Life Science, Guizhou University, Guiyang, China
[2]Research and Development Center of Traditional Chinese Medicinal and Natural Products, Guizhou University, Guiyang, China
[3]Guizhou Engineering Laboratory of Propagation and Cultivation on Traditional Chinese Medicinal Materials, Guiyang, China
Email: sci.blin@gzu.edu.cn

ABSTRACT

The drying process of traditional Chinese medicine *Rosa laevigata* Michx was investigated in a laboratory scale dryer. Drying experiments were conducted at five temperatures of 40°C, 50°C, 60°C, 70°C and 80°C. The experimental data were fitted to ten different thin layer drying models to select a suitable model for describing the drying process of *Rosa laevigata* Michx fruits. The results showed that the Midilli model had the highest value of R^2 (0.999767), the lowest χ^2 (0.000034) and *RMSE* (0.003616) among the ten models considered. The drying process of *Rosa laevigata* Michx fruits consisted of 1st falling rate period and 2nd falling rate period. The effective moisture diffusivity was found in the range of 1.49 - 30.20 × 10^{-11} $m^2 \cdot s^{-1}$. The value of activation energy obtained by plotting $\ln D$ versus $1/T$ is about 36 kJ·mol^{-1}.

Keywords: Drying; Mathematical Modelling; Kinetics; *Rosa laevigata* Michx

1. Introduction

It is well known that traditional Chinese medicine (TCM) has been used in the therapy of many diseases for several thousand years in China and neighbor countries and plays a more and more important role in modern pharmaceutical industry. In the process of refining TCM, drying is the most important methods and the oldest ways to remove the moisture in the solid. The main objective in drying herbal medicine is to minimize biochemical, chemical and microbiological deterioration of herbal medicine due to the reduction of the moisture content to a certain level, which allows safe storage over a long period and bring substantial reduction in weight and volume, minimizing packaging, storage and transportation costs [1]. Moisture content of TCM should be controlled under safe level according to Chinese Pharmacopoeia. *Rosa laevigata* Michx, an evergreen climbing shrub widely distributed throughout southern China, is a member of the Rosaceae family. The fruit of this plant is widely used as a traditional Chinese folk medicine and also used as a foodstuff in the traditional brewing of wine, as well as for the extraction of brown pigments used as food additives. In addition, *Rosa laevigata* Michx fruits are prescribed in the Chinese Pharmacopoeia for the treatment of wet dreams, urinary incontinence, urinary frequency, uterine prolapse, menstrual irregularities and leucorrhea [2]. *Rosa laevigata* Michx fruits contains polysaccharide, organic acid, tannins, triterpene, vitamin C and other components [3]. Polysaccharide is the main component and is used as quality assessment criteria, no less than 25% (w/w) according to Chinese pharmacopoeia [4]. There are two harvest periods for *Rosa laevigata* Michx fruits: the first is when it is ripe (usually from October to December), and the second is when it is partly yellow but not yet ripe (usually from August to September) [3].

Growing attentions have been paid to pharmacological effect of *Rosa laevigata* Michx on curing hyperpiesia, chronic cough, and dermatogic disease etc. [5]. However, systematic studies on the drying process of *Rosa laevigata* Michx fruits was not found as far as we known. So far, most of methods for drying TCM, which usually leads to high energy-consuming, time-consuming and degradation of biochemical active component, are empirical. In order to reduce energy consumption, save time and avoid degradation of biochemical active component, the drying model of TCM needs to be established by experimentation and verified by comparing simulated values with experimental data. The principle of modeling is based on having a set of mathematical equations that can adequately describe the system. The solution of these equations must allow prediction of parameters, such as moisture content, energy consumption etc., as a function of time at any point in the dryer based only on the initial conditions. Therefore the use of a simulation model is a valuable tool for prediction of drying time and optimization of dryers [6]. The aim of this work is to 1) evaluate the suitability of the drying models widely used to describe agricultural products for the drying process of tra-

ditional Chinese medicine; 2) study the drying kinetics and determine the moisture diffusivity and activation energy during drying of *Rosa laevigata* Michx fruits.

2. Materials and Methods

2.1. Samples Preparation

Fresh *Rosa laevigata* Michx fruits were collected from Guiyang, Guizhou Province, China, in November 2011. Then the fruits were cleaned and stored in a refrigerator at 4°C ± 1°C. At the start of each experiment, the fruits were hemisected and separated from the seeds according to Chinese Pharmacopoeia.

2.2. Instruments

The drying experiments were carried out using a laboratory dryer (DGG-9246A type, Shanghai Keelrein Instrument Co. Ltd.) in technical center of Guizhou University & Guizhou Bailing Group Pharmaceutical Company Limited. The dryer had an automatic temperature controller and is capable of providing any desired drying air temperature in the range of RT (room temperature) +10 to 300°C with an accuracy of ±0.1°C. The temperature and relative humidity of the room where the dryer was installed was about 10°C - 20°C and 40% - 65% respectively. Prior to placing the sample into the drying chamber, the system had been running for at least 0.5 h to obtain steady conditions.

The moisture content (dry basis, kg water/kg solids) of the *Rosa laevigata* Michx fruits was measured by Mettler Toledo HR83 halogen moisture analyzer. The halogen moisture analyzer provides fast and reproducible results with repeatability of 0.05% (2 g sample) moisture content. The temperature range that the halogen moisture analyzer provided is between 40°C to 200°C. Substances which subject to infrared ray with very low moisture content (10 ppm) can be analyzed by this instrument.

2.3. Drying Experiments

The drying experiments were conducted at five temperatures of 40°C, 50°C, 60°C, 70°C and 80°C. About 120 g of *Rosa laevigata* Michx fruits were uniformly spread in a tray and kept inside the dryer. About 15 g fruits were taken out of the dryer at each suitable interval and then grinded to powder by an electric grinder. The moisture content of the fruits powder was measured by the halogen moisture analyzer at 105°C with standard temperature program. Each drying experiment was repeated three times.

3. Mathematical Modeling

The moisture ratio (*MR*) of *Rosa laevigata* Michx fruits

during experiments was calculated by the following equation:

$$MR = \frac{M_t - M_e}{M_0 - M_e} \tag{1}$$

where M_t is the moisture content at time t, M_0 and M_e are the initial and equilibrium moisture contents, respectively, on dry basis. Since the values of M_e are relatively little, Equation (1) can be simplified as [6]:

$$MR = \frac{M_t}{M_0} \tag{2}$$

To select a suitable model for describing the drying process of *Rosa laevigata* Michx fruits, drying curves were fitted to ten well known thin layer drying models [7-13] which are presented in **Table 1**.

The non-linear regression analysis was done using IBM SPSS version 19.0. Three parameters, coefficient of determination (R^2), reduced chi-square (χ^2) and root mean square error (*RMSE*) were used as criteria to select the best equation to account for drying curves. These statistical values can be calculated as follows [6,12]:

$$R^2 = 1 - \left[\frac{\sum_{i=1}^{N} \left(MR_{exp,i} - MR_{pre,i} \right)^2}{\sum_{i=1}^{N} \left(MR_{exp,i} - \overline{MR}_{pre,i} \right)^2} \right] \tag{3}$$

$$\chi^2 = \frac{\sum_{i=1}^{N} \left(MR_{exp,i} - MR_{pre,i} \right)^2}{N - n} \tag{4}$$

$$RMSE = \left[\frac{1}{N} \sum_{i=1}^{N} \left(MR_{exp,i} - MR_{pre,i} \right)^2 \right]^{1/2} \tag{5}$$

where $MR_{exp,i}$ is the ith experimental moisture ratio, $MR_{pre,i}$ is the ith predicted moisture ratio, N is the number of observations and n is the number of parameters in the equations that has been used. The best model describing the thin layer drying characteristics of *Rosa laevigata* Michx was chosen as the one with the highest R^2-value and the lowest *RMSE* and χ^2-values.

Since the thickness of hemisected fruits of *Rosa laevigata* Michx was about 2 mm, the equation of Fick's second law was used for calculation of effective moisture diffusivity of drying process. Fick's diffusion equation was expressed as follows [6]:

$$MR = \frac{M_t}{M_0} = \frac{8}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{(2n-1)^2} \exp\left[-\frac{(2n-1)^2 \pi^2 Dt}{4L^2} \right] \tag{6}$$

where $n = 1, 2, 3, \cdots$ the number of terms taken into consideration, t is the time of drying (s), D is effective moisture diffusivity ($m^2 \cdot s^{-1}$) and L is the thickness of slice (m). Only the first term of Equation (6) is used for long time drying [14]:

Table 1. Thin layer drying models applied to drying curves.

Model No.	Model name	Equation
1	Newton	$MR = \exp(-kt)$
2	Page	$MR = \exp(-kt^n)$
3	Henderson and Pabis	$MR = a\exp(-kt)$
4	Aghbashlo *et al.*	$MR = \exp[-k_1 t/(1+k_2 t)]$
5	Logarithmic	$MR = a\exp(-kt)+c$
6	Two term	$MR = a\exp(-k_0 t)+b\exp(-k_1 t)$
7	Two-term exponential	$MR = a\exp(-kt)+(1-a)\exp(-kat)$
8	Wang and Singh	$MR = 1+at+bt^2$
9	Diffusion approach	$MR = a\exp(-kt)+(1-a)\exp(-kbt)$
10	Midilli *et al.*	$MR = a\exp(-kt^n)+bt$

$$MR = \frac{M_t}{M_0} = \frac{8}{\pi^2} \exp\left[-\frac{\pi^2 D t}{4L^2}\right] \quad (7)$$

Taking the natural logarithm on both sides of Equation (7) yields:

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

According to Equation (8), a straight line can be obtained by plotting ln*MR* versus t. Thus, the effective moisture diffusivity *D* can be determined from the slope. The relationship between effective moisture diffusivity *D* and temperature is represented by an Arrhenius type equation [14]:

$$D = D_0 \exp\left(-\frac{E_a}{RT}\right) \quad (9)$$

where D_0 is the pre-exponential factor of the Arrhenius equation ($m^2 \cdot s^{-1}$), E_a is the activation energy ($kJ \cdot mol^{-1}$), R is universal gas constant ($8.314 \ kJ \cdot mol^{-1} \cdot K^{-1}$) and T is absolute temperature (K). Equation (9) can be linearized by taking natural logarithm on both sides:

$$\ln D = \ln D_0 - \frac{E_a}{RT} \quad (10)$$

The activation energy can be calculated from the slope of the plot ln*D* versus $1/T$.

4. Results and Discussion

The initial moisture content of the raw fruits was 193.00 ± 2.05 kg/kg (dry basis). The drying curves of *Rosa laevigata* Michx fruits at 40˚C, 50˚C, 60˚C, 70˚C and 80˚C are shown in **Figure 1**.

The final moisture content of *Rosa laevigata* Michx fruits ranged from 5.72% to 10.85% (dry basis). As can
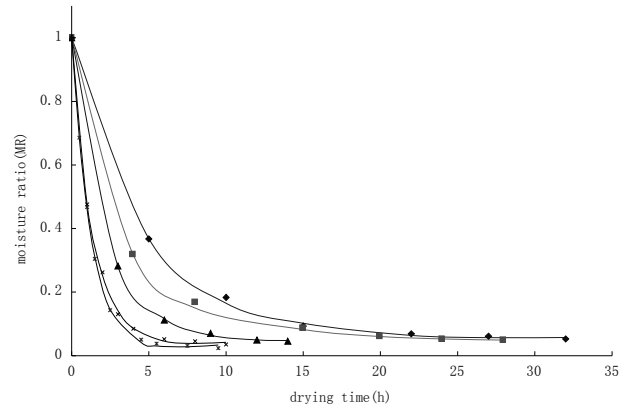
Figure 1. Drying curves of *Rosa laevigata* Michx fruits at 40˚C, 50˚C, 60˚C, 70˚C and 80˚C. Experimental data and values calculated by the new model. Experimental: ◆: 40˚C; ■: 50˚C; ▲: 60˚C; ×: 70˚C; *: 80˚C. Simulated: (a) 40˚C; (b) 50˚C; (c) 60˚C; (d) 70˚C; (e) 80˚C.

be seen from **Figure 1**, the moisture ratios during the drying decreased exponentially as the drying time increased. It was evident that drying temperature significantly affected the drying process. The drying time reduced when the temperature increased. In another word, the drying rate increased with the increase in drying temperature.

The average moisture ratio of *Rosa laevigata* Michx fruits dried at different temperatures was fitted to ten different drying models. The statistical results from ten models are given in **Table 2**.

It was seen from **Table 2**. that the values of R^2 for the Aghbashlo model, Logarithmic model, Two term model, Diffusion approach model and Midilli model were greater than 0.999, indicating good fit. The Midilli model gave the highest value of $R^2 = 0.999767$, the lowest value of $\chi^2 = 0.000034$ and *RMSE* = 0.003616. Thus the Midilli model was comparatively the better drying model to represent the drying process of *Rosa laevigata* Michx fruits. **Figure 1** was the comparison of experimental *MR* and simulated *MR* obtained by the Midilli model versus drying time under different drying temperature, which indicated a good fit.

Based on Equation (8), the plots of ln*MR* versus drying time t at different temperatures were shown in **Figure 2**.

As can be seen from **Figure 2**, the constant rate period was absent, and the drying process of *Rosa laevigata* Michx fruits at different temperatures consisted of two regions, 1st falling rate period of drying and 2nd falling rate period of drying, having different slopes. The critical moisture content represents of the time when the surface was completely dried and moisture was transported from inside of the fruits to the surface by capillary action [15]. Similar situation were observed by C. T. Akanbi [16], and T. Arumuganathan [12] etc. The best-fit regression

Table 2. Statistical results of the drying model.

Model name	R^2	χ^2	*RMSE*
Newton	0.988974	0.001300	0.030751
Page	0.998097	0.000174	0.009800
Henderson and Pabis	0.989092	0.001549	0.030584
Aghbashlo *et al.*	0.999165	0.000084	0.007132
Logarithmic	0.999183	0.000128	0.007337
Two term	0.999687	0.000054	0.004217
Two-term exponential	0.993882	0.022797	0.011560
Wang and Singh	0.864878	0.016367	0.102000
Diffusion approach	0.999681	0.000041	0.004226
Midilli *et al.*	0.999767	0.000034	0.003616
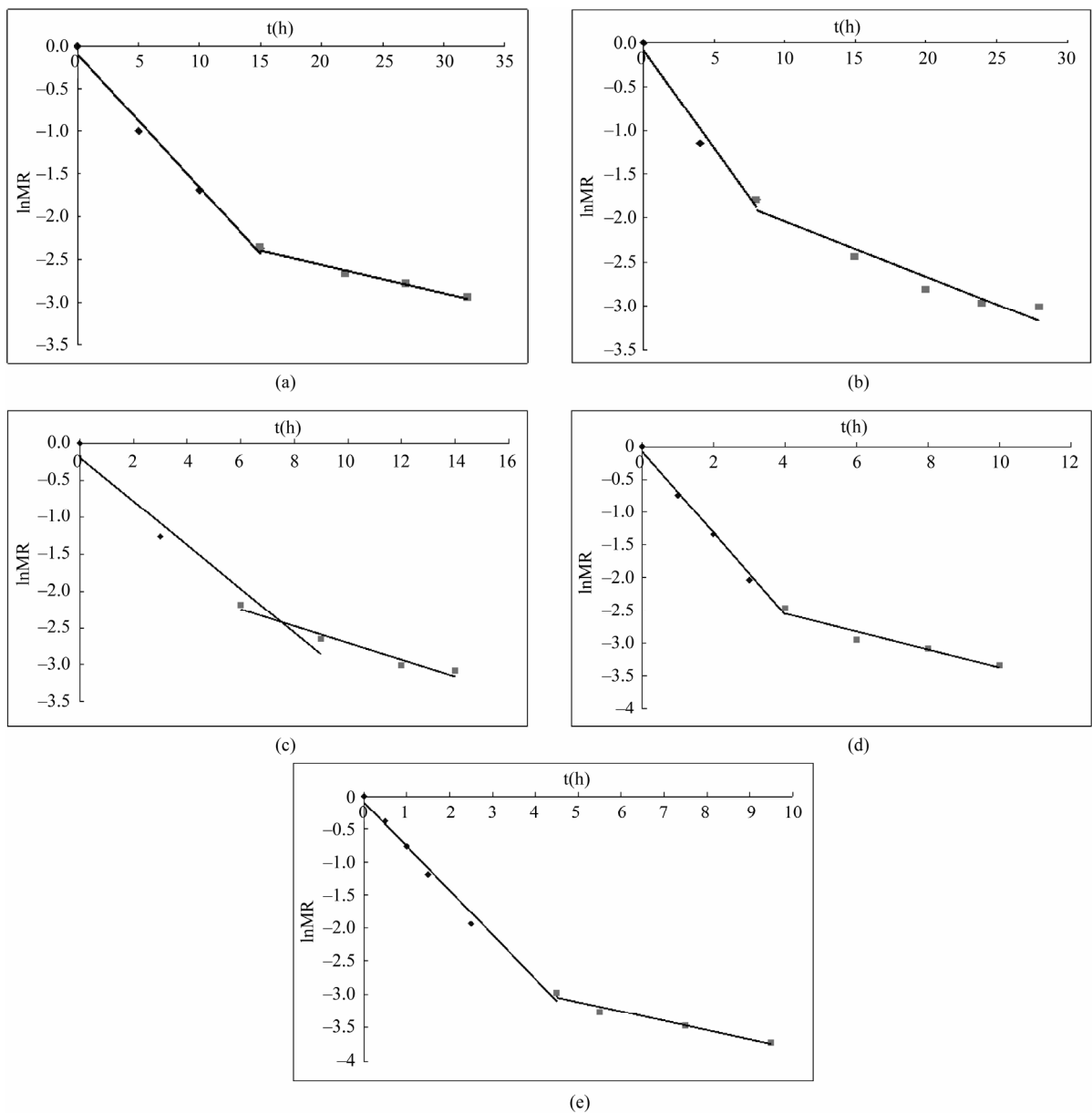
Figure 2. The plots of ln*MR* versus drying time t for drying of *Rosa laevigata* Michx fruits at temperatures. (a) 40˚C; (b) 50˚C; (c) 60˚C; (d) 70˚C and (e) 80˚C.

equations at different temperature during first and second falling rate periods of drying were given in **Table 3**.

It was found that during the first and second falling rate periods the absolute values of slop of regression lines were increased with the increase of temperature. The values of first and second effective moisture diffusivity D at different temperatures were calculated from the slops of regression lines according to Equation (8) and listed in **Table 4**.

It was observed from **Table 4**, as expected, the drying temperature affect the values of D greatly. Effective moisture diffusivity values of first and second falling rate increase with the increase in drying temperature. These values were within the general range of 10^{-9} - 10^{-11} $m^2 \cdot s^{-1}$ for agricultural products [17]. Two straight lines were obtained by plotting $\ln D$ versus $1/T$ with correlation coefficient R^2 0.9672 and 0.9300 respectively (**Figure 3**). Therefore, the values of activation energy could be calculated from the slope of the straight lines (also listed in **Table 4**).

Figue 3 showed the variation of activation energy of 1st falling rate period and that of 2nd falling rate period

was small. The effect of temperature on Effective moisture diffusivity during 1st falling rate period was consistent with that during 2nd falling rate period. The value of activation energy obtained was about 36 kJ/mol which was in the general range of 12.7 - 110 $kJ \cdot mol^{-1}$ for agricultural products [18].

5. Conclusion

Mathematical modelling of the drying process was used to agricultural products widely [7,9-13], but not yet for traditional Chinese medicine. In this work, the drying process of *Rosa laevigata* Michx fruits was studied. The results of model simulation indicated the Midilli model gave the best results and showed good agreement with the experimental values of drying of *Rosa laevigata* Michx fruits. The values of drying kinetics parameters effective moisture diffusivity D and activation energy E_a which obtained through Fick's second law and Arrhenius equation were within the general range of 10^{-9} - 10^{-11} $m^2 \cdot s^{-1}$ and 12.7 - 110 $kJ \cdot mol^{-1}$ respectively. Hence, mathematical modelling could be an effective tool for the

Table 3. Regression equations and correlation coefficients of 1st and 2nd falling rate of drying.

Drying temperature (°C)	1st falling rate of drying		2nd falling rate of drying	
	Regression equation	R^2	Regression equation	R^2
40	$y = -0.1563x - 0.0952$	0.9902	$y = -0.0330x - 1.9045$	0.9840
50	$y = -0.2234x - 0.0847$	0.9737	$y = -0.0626x - 1.4148$	0.9277
60	$y = -0.2960x - 0.1942$	0.9608	$y = -0.1145x - 1.5583$	0.9614
70	$y = -0.5820x - 0.0714$	0.9933	$y = -0.1384x - 1.9934$	0.9402
80	$y = -0.6699x - 0.0926$	0.9892	$y = -0.1592x - 2.4238$	0.9632

Table 4. Effective moisture diffusivity D and activation energy E_a for drying of *Rosa laevigata* Michx.

	$D \times 10^{-11}$ ($m^2 \cdot s^{-1}$)					E_a ($kJ \cdot mol^{-1}$)
	40°C	50°C	60°C	70°C	80°C	
1st falling rate of drying	7.05	10.07	13.34	26.24	30.20	35.51
2nd falling rate of drying	1.49	2.82	5.16	6.24	7.18	36.56
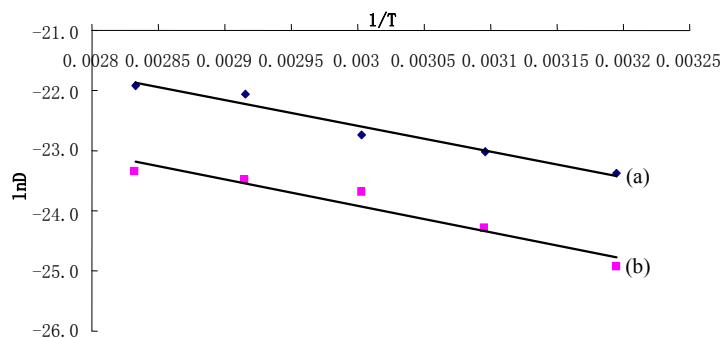
Figure 3. The plots of $\ln D$ versus $1/T$ for drying of *Rosa laevigata* Michx fruits. (a) 1st falling rate; (b) 2nd falling rate.

study of drying behavior of traditional Chinese medicine. The suitability of mathematical modelling for the drying process of other traditional Chinese medicine should be study in other works.

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