Hot-air Drying of Rehmannia Root: Its Kinetic Parameter, Shrinkage and Mathematical Modelling

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Abstract

Rehmannia root has been used as an herbal medicine and health food for more than two thousand years in many Asian countries. To improve the efficiency of root drying process, fresh root was dried at 70, 80, 90 and 100°C, and an air velocity of 0.5 m·s-1 in a laboratory dryer. The drying was found to be a typical falling rate drying all the time except the initial, and substantially influenced by air temperature. Among 12 mathematical models, logarithmic equation best described the hot-air drying of rehmannia root. The relationship between drying constant (k) in logarithmic equation and drying temperature met Arrhenius equation, and the activation energy of rehmannia root drying was estimated to be 26.75 kJ·mol-1. Volume shrinkage linearly correlated with mass loss, however, at the end of rehmannia root drying, volume expansion was detected, especially at a higher temperature. By plotting apparent density versus moisture ratio (MR), two turning points of volume alteration were identified (MR = 0.1 and 0.4). These findings will be of great interest for the industrial drying of rehmannia root.

Keywords: *Rehmannia glutinosa*; mathematical modeling; volume shrinkage, hot-air drying, activation energy

1. Introduction

Rehmannia glutinosa (Gaertn.) DC. is a perennial of family Plantaginaceae. Its tuberous root is one of the most widely used medicinal herbs and health foods in East Asian countries, including China, Korea, and Japan etc. China is the main producer of rehmannia root, and its annual output is about 25,000 tons (1-4).

Fresh rehmannia root contains a large amount of water and is liable to rot. Traditionally, the dried rehmannia root is the main form for preservation and delivery, and it is also the raw material to make slices ready for decoction and to prepare steamed rehmannia root. In the harvest season of rehmannia root, late autumn, it is difficult to dry the root by sun. The fresh root is usually dried in whole by open fire. In recent years, modern drying methods have been frequently tried. Liu et al (5) and Liu et al (6) reported vacuum infrared radiation drying of rehmannia root, and its mathematical modelling, mass and heat transfer analysis. Rhim et al (7) and Lu et al (8) reported the desorption isotherm of rehmannia root. The hot air drying kinetic parameters of *R. glutinosa* var. *purpurea* were also determined by Rhim et al (7). The other reports, however, mainly related different drying methods with the quality of the products (3, 9-10).

Rehmannia glutinosa 'Beijing 3' is the main cultivar in production, however, no information is available in literature on its drying characteristics. In addition, there exists a controversy between early drying at a high temperature and then at a low temperature and the reverse. The objective of this study was to determine the drying characteristics of rehmannia root, focusing on drying kinetic parameters, volume shrinkage, and mathematical modeling.

2. Materials and Methods

2.1 Experimental material

Fresh rehmannia root (*Rehmannia glutinosa* 'Beijing 3') was harvested from Dongping County, Shandong Province, China. Homogenous roots (50-60 mm in diameter) was washed with tap water to remove attached soil on the surface and stored in polyethylene bags at 4°C until drying experiments. The initial moisture of fresh root was found to be 75.06% (wet basis), determined by drying the slices (1-2 mm thick) of the root at 105°C.

2.2 Drying procedure

The drying experiment was conducted in a laboratory dryer with forced convection. Five fresh rehmannia roots (460-480 g), after 4 h stabilization at ambient temperature (20-25°C), were put on a stainless steel mesh (25×25 cm), and dried at temperatures of 70, 80, 90 or 100°C and an air velocity of 0.5 $m\cdot s^{-1}$. Volume change and moisture loss were recorded at a 2-h interval during the drying process. Drying was continued until no further weight change (about 25-27%, wet basis). Each drying has six duplicates to obtain a reasonable average.

2.3 Volume and mass determination

The volume of the tested roots was determined by a volume displacement method. Briefly, the 5 tested roots were put into a 1000 ml measuring cylinder, in the same time, fine sand (about 100 μm in diameter) at the same temperature was poured into the vessel and added to the scale after vibration for 10 s, then poured out, and the sand volume was measured again, their difference was the volume of the roots. The mass measurements were done simultaneously with an electronic balance with a sensitivity of 0.01 g.

2.4 Data processing

2.4.1 Moisture content and drying rate

In drying experiments, the moisture content of tested material usually expressed in dimensionless form as moisture ratio (M_R), which was calculated using Eq. 1:

$$M_R = \frac{M_t - M_0}{M_0 - M_e} \qquad (1)$$

Where: M_t, M_0 and M_e were the moisture content (kg water·kg^{-1} dry matter) at time t, initial and equilibrium moisture content, respectively. The value of M_e is relatively small compared to M_t or M_0; hence, the error involved in the simplification is negligible. Thus, the moisture ratio was simplified to Eq. 2 (11-13):

$$M_R = M_t / M_0 \qquad (2)$$

In addition, drying rate was calculated as moisture removed per unit time and per unit dry matter (kg water·kg^{-1} dry matter·h^{-1}).

2.4.2 Mathematical modelling of rehmannia root drying

The drying experimental data of rehmannia root were fitted to twelve commonly used thin layer drying models (Table 1), using non-linear regression solved by a Levenberg-Marquardt numerical algorithm. The fitness of the tested mathematical models to the experimental data was evaluated with 4 parameters, the coefficient of determination (R^2), the reduced chi-square (χ^2), the root mean square error ($RMSE$) and the mean relative percent error modulus (P). The higher the values of R^2 and the lower the values of P, $RMSE$ and χ^2, the better the goodness of the fitting (14-15). These parameters were calculated as Eqs. (3), (4), (5) and (6), respectively:

$$R^2 = 1 - \frac{\sum_{i=1}^{N} \left(MR_{\exp,i} - MR_{pre,i}\right)^2}{\sum_{i=1}^{N} (MR_{\exp,i} - MR_{premean})^2} \qquad (3)$$

$$\chi^2 = \frac{\sum_{i=1}^{N} (MR_{\exp,i} - MR_{pre,i})^2}{N - z} \qquad (4)$$

$$RMSE = \sqrt{\frac{\sum_{i=1}^{N} (MR_{\exp,i} - MR_{pre,i})^2}{N}} \qquad (5)$$

$$P = \frac{100}{N} \sum_{i=1}^{N} \frac{\left|MR_{\exp,i} - MR_{pre,i}\right|}{MR_{\exp,i}} \qquad (6)$$

Where: $M_{R\ exp,i}$ is the experimental moisture ratio at time t, $M_{R\ pre,i}$ is the corresponding moisture ratio predicted by a tested model, $M_{R\ pre,mean}$ is the mean moisture ratio predicted by a tested model, N is the number of observations and z is the number of constants.

2.4.3 Calculation of activation energy

In the falling rate drying, diffusion plays dominant role in moisture movement within the material, which can be described by the Fick's second law of diffusion. The simplified solution of diffusion equation is usually applied to calculate effective diffusivity, which is necessary to obtain activation energy based on Arrhenius equation. Unfortunately, the rehmannia root is irregular in shape, thus the method could not be applied. Previous studies have found that one parameter of drying models (Table 1), drying constant (k), was also fit the Arrhenius equation. The values of activation energy obtained from drying kinetics data were very close to those from diffusivity data (7, 16), therefore, the activation energy of rehmannia root was estimated according to Eq. 7:

$$k = A \exp\left(\frac{-E_a}{RT}\right) \qquad (7)$$

Where A is the pre-exponential factor, E_a is the activation energy (kJ·mol^{-1}), T is the absolute temperature (K) and R is the universal gas constant (8.314×10^{-3} kJ·mol^{-1}·K^{-1}).

2.4.4 Shrinkage of rehmannia root during hot air drying

Shrinkage is a common physical phenomenon of biomaterial drying, leading to change in organoleptical, textural and rehydration properties of the dried products, especially in fruits and vegetables. Many models have been developed to describe the shrinkage, including empirical and fundamental fitting of the experimental shrinkage data as a function of moisture content or moisture ratio (17-19). To describe the shrinkage of rehmannia root, volume ratio (V_R) and apparent density (ρ_t) at a given time, were calculated according to Eqs. (8) and (9), respectively:

$$V_R = V_t / V_0 \quad (8)$$

$$\rho_t = W_t / V_t \quad (9)$$

Where V_0 is the initial volume, V_t *and* W_t are the volume and the weight of the material at a given time *t*.

Since the more the water removed the more contraction stresses are originated in the material, at a relative low drying rate, shrinkage of the material ideally equals the volume of removed water, therefore, it can be described by the linear empirical model as Eq. 10 (17-19):

$$V_R = aM_t + b \quad (10)$$

Where *a* and *b* were the coefficient and the constant of the model, respectively.

3. Results and Discussion

3.1 Drying characteristics of rehmannia root

The drying curves and drying rates of rehmannia root are shown in Fig. 1 and 2. It is clear that the drying displays the characteristics of a falling rate drying. The drying from the initial moisture content of 3.0095 to 0.1000 kg water·kg^{-1} dry matter took 54, 52, 32 and 22 h at 70, 80, 90 and 100°C, respectively. The result indicates that drying temperature markedly influence the efficiency of rehmannia root drying as widely observed in foodstuff dryings (7, 11, 13, 15, and 19).

At the beginning of the dryings, an increasing rate drying was detected, which might be due to high energy needed to heat the material. The result was in agreement with the previous report of Rhim et al. (7). Similar results were also reported for strawberry (11), potato slices (19).

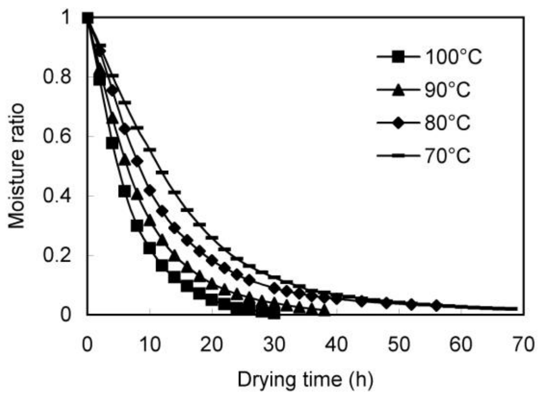

Fig. 1. Drying curves of rehmannia root at different temperatures.

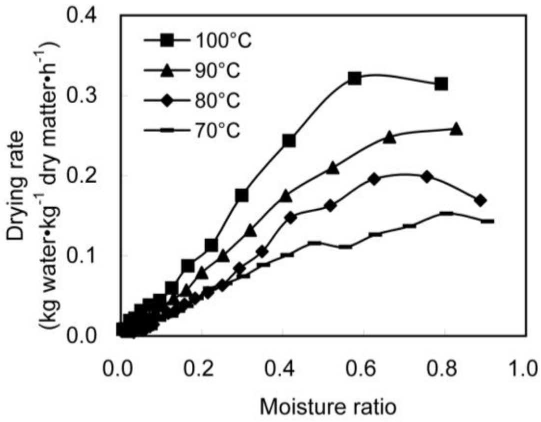

Fig. 2. Drying rates of rehmannia root at different drying temperatures.

3.2 Mathematical modelling of rehmannia root drying

Experimental data of rehmannia root dryings at 70, 80, 90 and 100°C were converted into moisture ratios and fitted to twelve models commonly used in thin layer convective dryings. The calculated values of R^2, χ^2, *RMSE* and *P* are summarized in Table 1. The values of R^2 are larger than 0.9960, those of χ^2 and *RMSE* less than 0.0005 and 0.0200, respectively, for all but Wang and Singh model. The variation range of *P* values is relatively larger, ranging from 1.82% to 184.44%. According to Madamba et al. (14) and Özdemir & Devres (15), a *P* value lower than 10% is recommended for the selection of models. Taking the four assessment criteria, logarithmic model and approximation of diffusion model is comparable, therefore, the former is selected to describe the convective drying of rehmannia root for its simpleness. Its fitness is shown in Fig. 3. In literature, this model is also successfully used to describe the drying behaviors of strawberry (11), sour cherry (20) etc.

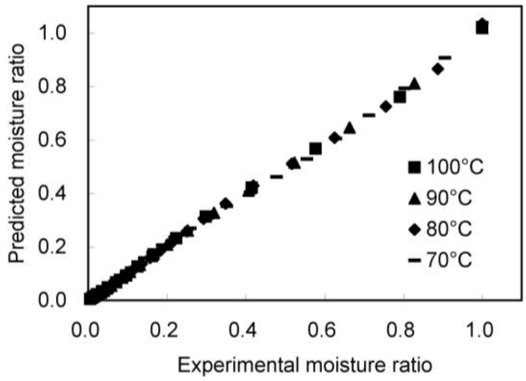

Fig. 3. Comparison of the experimentally obtained moisture ratios and the logarithmic model predicted moisture ratios for convective air drying of rehmannia root.

Table 1 Statistical criteria of various tested models for rehmannia root drying at different drying temperatures

Model name and mathematical expression [a]	Drying temperatures (°C)	R^2	χ^2	*PMSE*	*P*
Lewis model	70	0.9966	0.0003	0.0161	11.46
$M_R = \exp(-kt)$	80	0.9959	0.0003	0.0179	16.21
	90	0.9989	0.0001	0.0096	4.72
	100	0.9979	0.0002	0.0134	20.11
Henderson and Pabis model	70	0.9978	0.0002	0.0128	12.11
$M_R = a\exp(-kt)$	80	0.9964	0.0003	0.0168	17.37
	90	0.9964	0.0003	0.0168	17.37
	100	0.9985	0.0001	0.0114	16.56
Logarithmic model	70	0.9978	0.0002	0.0128	10.85
$M_R = a\exp(-kt) + c$	80	0.9981	0.0002	0.0121	3.16
	90	0.9992	0.0001	0.0080	3.29
	100	0.9987	0.0001	0.0107	4.59
Page model	70	0.9987	0.0001	0.0100	18.17
$M_R = \exp(-kt^n)$	80	0.9960	0.0003	0.0177	17.67
	90	0.9993	0.0001	0.0075	9.69
	100	0.9992	0.0001	0.0081	7.74
Wang and Singh model	70	0.9565	0.0035	0.0575	84.76
$M_R = 1 + at + bt^2$	80	0.9303	0.0060	0.0742	70.13
	90	0.9576	0.0038	0.0584	83.80
	100	0.9577	0.0042	0.0604	184.44
Midilli model	70	0.9998	0.0000	0.0042	5.94
$M_R = a\exp(-kt^n) + bt$	80	0.9989	0.0001	0.0092	6.82
	90	0.9997	0.0000	0.0049	6.97
	100	0.9993	0.0001	0.0077	15.12
Two term model	70	0.9980	0.0002	0.0123	6.47
$M_R = a\exp(-k_0t) + b\exp(-k_1t)$	80	0.9982	0.0002	0.0118	3.98
	90	0.9992	0.0001	0.0079	4.00
	100	0.9985	0.0002	0.0116	18.68
Two term exponential model	70	0.9986	0.0001	0.0103	19.15
$M_R = a\exp(-kt) + (1-a)\exp(-akt)$	80	0.9962	0.0003	0.0173	12.68
	90	0.9991	0.0001	0.0085	10.12
	100	0.9990	0.0001	0.0091	8.15
Modified Henderson and Pabis model	70	0.9979	0.0002	0.0125	14.87
$M_R = a\exp(-kt) + b\exp(-gt) + c\exp(-ht)$	80	0.9999	0.0000	0.0027	1.82
	90	0.9992	0.0001	0.0081	5.85
	100	0.9985	0.0002	0.0114	16.56
Approximation of diffusion model	70	0.9966	0.0003	0.0159	7.63
$M_R = a\exp(-kt) + (1-a)\exp(-bkt)$	80	0.9972	0.0002	0.0148	5.25
	90	0.9989	0.0001	0.0095	3.85
	100	0.9985	0.0002	0.0114	5.63
Verm model	70	0.9992	0.0001	0.0080	14.88
$M_R = a\exp(-kt) + (1-a)\exp(-gt)$	80	0.9972	0.0002	0.0148	5.25
	90	0.9989	0.0001	0.0095	3.85
	100	0.9985	0.0002	0.0114	5.63
Weibull distribution model	70	0.9998	0.0000	0.0036	4.60
$M_R = a - b\exp(-gt^n)$	80	0.9993	0.0001	0.0076	5.41
	90	0.9998	0.0000	0.0044	6.72
	100	0.9994	0.0001	0.0075	17.11

[a] M_R, moisture rates; *a*, *b* and *c*, drying coefficient specific to each model; *k*, k_0, k_1, g, h drying constants; *t*, drying time; *n*, exponent

3.3 Activation energy estimation

Making a plot of the natural logarithm of *k*, the drying constant of logarithmic model, as a function of the reciprocal of drying temperature (1/T) gave a straight line (R^2 = 0.9978) as presented in Fig. 4. The result indicates that the temperature dependence of *k* follows the Arrhenius equation. Based on this relationship, the activation energy was calculated to be 26.75 kJ·mol^{-1}, which is much lower than the value (38.26 kJ·mol^{-1}) reported by Rhim et al. (7) with the same method for *R. glutinosa* var. *purpurea*. The deviation may be due to the genetic difference of the two samples, since the diameter of our sample is about 4-5 times as large as that of Rhim et al. (7).

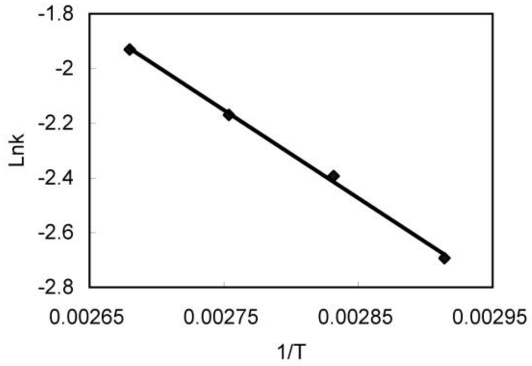

Fig. 4. Arrhenius-type relationship between drying constant in logarithmic model and drying temperatures.

3.4 Volume shrinkage

Like many foods and vegetables with high water content, volume reduction of rehmannia root upon hot air drying is conspicuous as shown in Fig. 5. When the sample volume reached 24.6%, 24.7% and 32.5% of the initial volume in the dryings at temperatures of 80, 90 and 100°C, respectively, the samples began to expand, the phenomenon was not obvious at 70°C drying. This final expansion has not been reported in the dryings of foodstuffs (18). The reason may be due to the property of the tested material itself.

The rehmannia root is a storage organ, having abundant parenchyma and less sclerenchyma, its mainly storage nutrient is stachyose, a tetrasaccharide (about 50% of its dry matter) (21). These characteristics make it easier to collapse upon water removal and render its matrix high mobility. During the early stage of drying, sample shrinkage prevailed, at the end of drying, however, the surface became drier than the centre, making the surface stiff and limiting shrinkage. When drying at a higher temperature, low moisture shell or crust formed during the early drying might reduce water diffusion and increase inner stresses, in the meantime, viscosity increased with the rise of material temperature. The combined effects of these factors should be responsible for the expansion phenomenon. In contrast, at a lower drying temperature, such as 70°C, drying rate was low, moisture profile in the sample was relatively flat, and internal stress might be minimum, therefore, the root expansion was nearly unnoticed, as shown in Fig. 5.

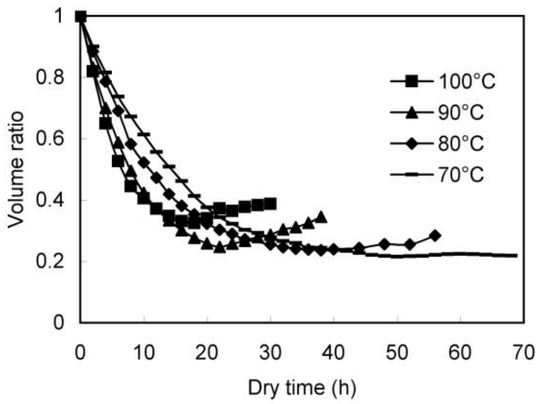

Fig. 5. Volume shrinkage as a function of drying time for rehmannia root drying at different temperatures.

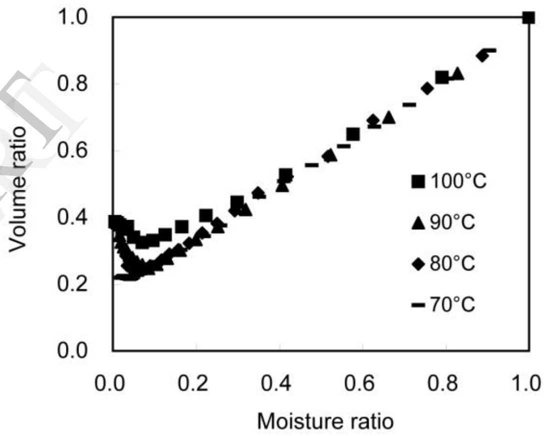

Fig. 6. Volume shrinkage as a function of moisture ratio for rehmannia root drying at different temperatures.

Volume shrinkage curves of rehmannia root are illustrated in Fig. 6. The curves are linear and overlap at the moisture ratio from 1.0 to 0.1 for the dryings at 70, 80 and 90°C, which can be described by Eq. 11:

$$V_R = 0.81M_R + 0.1732,\ R^2 = 0.9987 \qquad (11)$$

The result indicates that the volume reduction is strictly proportional to mass loss. The shrinkage at this stage should be regarded as ideal. At the end stage, the curves deviate from the above fitted straight line as shown in Fig. 6. This phenomenon has also been observed in several foodstuffs, such as potato, sweet potato, apple etc. (22-24). As to the drying at 100°C, the shrinkage behavior altered more early than that of dryings at other tested temperatures, and can be divided into three linear stages and described by Eqs. 12, 13, and 14, respectively:

$M_R > 0.4$:

$$V_R = 0.8089M_R + 0.1861,\ R^2 = 0.9992 \quad (12)$$

$0.1 < M_R < 0.4$:

$$V_R = 0.5677M_R + 0.2770,\ R^2 = 0.9993 \quad (13)$$

$M_R < 0.1$:

$$V_R = -\ 0.9551M_R + 0.3932,\ R^2 = 0.9236 \quad (14)$$

At the first stage of 100°C drying ($M_R > 0.4$), the curve nearly overlaps with those of the other temperatures (Eq.11 and 12). When the drying proceeded into the second stage, the shrinkage became slow, if not due to sudden surface hardening then some important changes probably happened. When converting our data into apparent density, the turning points are displayed more distinctly as shown in Fig. 7. We consider that the thermal damage of cambium cells should be responsible for this arresting change at $M_R = 0.4$. Cambium cells are relatively small, lack of intercellular space, with dense protoplasm and few vacuoles. Living cambium cells can prevent the root shrinkage. Once this part was destructed, shrinkage or expansion would become more facile as shown in Fig. 7.

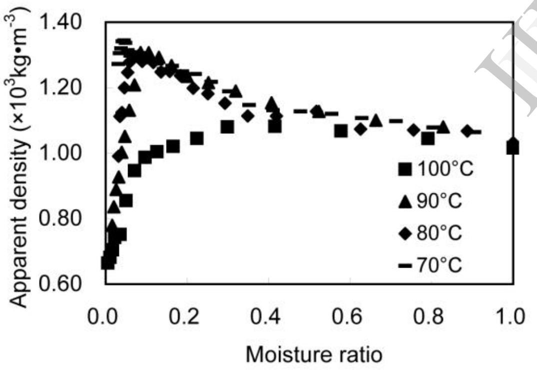

Fig. 7. Apparent density versus moisture ratio of rehmannia root drying at different temperatures.

Traditionally, the quality of dried rehmannia root is evaluated by weighing in hands or by water, the heavy or the submerged root is considered to be qualified. The exact mechanism for this assessment remains unknown. Several authors considered that the rehmannia root was deteriorated after hot air drying based on the decomposition of catalpol, one of the main secondary metabolites of *R. glutinosa* (9-10). However, our recent work verified that catalpol decomposition, especially its reaction with amino acids, led to substantial increase in antioxidant activity (25). Therefore, the effects of different drying methods should be reassessed by further pharmaceutical and clinical investigations.

Unlike the dryings of many foods and vegetables, shrinkage may be the desired character of rehmannia. Firstly, shrunk root can substantially save space for transport and storage. Secondly, the root having higher density than water will facilitate rehydration process before cutting operation for decoction. In view of the temperature dependence of moisture removal, shrinkage and final expansion, our results support the protocol of drying at a higher temperature early and then at a lower temperature, such as drying at 90°C and then at 70°C. This protocol can improve drying efficiency and avoid volume expansion.

4. Conclusion

The convection drying of rehmannia root was investigated in this study. It was found that the drying was a typical falling rate drying, the drying time significantly decreased with drying temperature increase. Logarithmic model could well describe the drying behavior of rehmannia root. The activation energy of the drying was estimated to be 26.75 $kJ{\cdot}mol^{-1}$ based on the Arrhenius type relationship between the drying constant in logarithmic model and drying temperature. Volume shrinkage of rehmannia root was found linearly correlated with mass loss in most drying times and drying temperatures. At the end of the dryings, a phenomenon of expansion was found. These findings were helpful to the improvement of industrial drying of rehmannia root.

Acknowledgements

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