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Dehydration Kinetics of Rehmannia (*Rehmannia glutinosa* Liboschitz)

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Abstract Sliced and whole root of rehmannia were dehydrated in a laboratory dryer at 40, 60, 80, and 100°C to evaluate the kinetic parameters for dehydration of rehmannia. The drying curves of both samples were characterized by a falling-rate drying period only. Sliced rehmannia dried 1.1 to 3.1 times faster than whole root of rehmannia depending on drying temperature. Equilibrium moisture content (EMC) of rehmannia samples at the drying temperature tested were 0.069-0.078 g water/g dry solid, which was coincided with the monolayer moisture content (0.06 and 0.07 g water/g dry solid) evaluated from desorption isotherms using GAB (Guggenheim-Anderson-de Boer) model. A logarithmic model for thin layer drying was applied to evaluate the drying time to reach EMC (t_{EMC}) and drying constant (k). The effect of temperature on $1/t_{EMC}$ and k was described by the Arrhenius model with activation energy values of 32.56 and 47.14 kJ/mol determined using the former parameter, and 34.27 and 38.26 kJ/mol determined using the latter parameter for sliced and whole root of rehmannia, respectively.

Keywords: rehmannia, *jiwhang*, dehydration, kinetics, logarithmic model

Introduction

Rehmannia, which is a tuber of a perennial herb of *Rehmannia glutinosa* Liboschitz var. *purpurea* Makino belonging to the family of *Scorophulariaceae* like ginseng, is one of the most widely used medicinal herb in the Oriental traditional medicine (1, 2). Known as Chinese foxglove, locally called *jiwhang* in Korea or *di huang* in China, rehmannia is often combined with other herbs to treat anemia, cancer, constipation, and diabetes. Other uses include the treatment of fatigue, high blood pressure, sleeping problems, and ringing in the ears. Rehmannia may also be used to treat skin diseases as well as to treat cuts and wounds. In the Oriental traditional medicine, rehmannia is used in three different types, *i.e.*, fresh (*Rehmanniae Radix crudus*; *saengjiwhang* or *sheng di huang*), dried (*R. Radix*; *gunjiwhang* or *sheng di huang*), and dried after steaming in rice wine (*R. Radix preparata*; *sookjiwhang* or *shu di huang*), depending on their preparation method and medicinal effect.

Usually, fresh rehmannia is a highly perishable and more than 30% of the crop is wasted due to weight loss and deterioration during storage (3). One of the ways to overcome this problem is to reduce water activity of rehmannia through the decrease in water content, avoiding potential deterioration and contamination during long storage periods, which is the most widely used method for preparation of dried rehmannia. Traditionally, dried rehmannia has been produced using a natural sun drying method. Although sun drying is the most commonly used method for the preparation of dried medicinal herb in the Oriental medicine, this technique is extremely weather dependent, and has the problem of contamination of foreign matter during prolonged drying period, consequently decrease in product quality. Therefore, applying modern drying technology, using drying methods such as hot air, vacuum, freeze drying, osmotic dehydration, vacuum impregnation methods, is required to produce high quality dried products.

There have been many studies on the dehydration characteristics of various vegetables such as red pepper (4-6), red chilli (7), garlic (8-10), carrot (11), okra (12), fig (13), mulberry (14), green bean (15), and green pea (16). Recently, demand on rehmannia including dried one is increasing with growing market in Korea (3), but little information on the dehydration characteristics and optimal drying conditions for the dehydration of rehmannia is available.

The main objective of this study was to determine the effect of drying temperature on drying kinetics of sliced and whole root of rehmannia.

Materials and Methods

Materials Fresh samples of rehmannia (*Rehmannia glutinosa* Liboschitz var. *purpurea* Makino) were obtained directly from a grower at Jangheung, Korea. The average initial moisture content of the rehmannia samples was 78.7± 0.6% (w.b.), as determined by vacuum drying at 70°C for 24 hr (17), and the average dimensions of the whole root of rehmannia were 16.4±5.6 cm in length and 1.2±0.2 cm in diameter. Before the drying process, samples were washed to remove dirt on the surface and drained to remove surface moisture. Samples were divided into 2 groups, one for the whole root of rehmannia and the other for sliced rehmannia. Sliced samples were obtained by cutting rehmannia roots into slices of 0.56±0.11 cm using a stainless steel knife.

Desorption isotherm Desorption isotherm of rehmannia samples were determined using a standard gravimetric method at 20°C with saturated salt solutions of known water activity (18) as shown in Table 1. About 10 g of sliced or 20 g of whole root of the rehmannia sample was

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Table 1. Saturated salt solution and equilibrium relative humidity (ERH) at 20°C

Saturated salt solution	ERH (%)	EMC (g water/g dry solid)	
		Sliced	Whole root
Lithium chloride	11.3	0.0715	0.0804
Magnesium chloride	33.1	0.0957	0.1141
Potassium carbonate	43.2	0.1191	0.1479
Magnesium nitrate	54.4	0.1465	0.1853
Sodium chloride	75.5	0.2602	0.3400
Potassium chloride	85.1	0.4671	0.9533
Potassium nitrate	94.6	2.4849	3.9711

taken into a sag made of nylon net and hanged inside a 2 L wide-necked glass jar saturated with saturated salt solution, and sealed by a screw collar and rubber gasket. Sample weight was measured periodically until a constant weight was reached. Once equilibrium was reached, the moisture content of the samples was determined according to the AOAC method (17). Then the equilibrium moisture content (EMC) of the rehmannia sample was plotted against water activity (a_w) of each saturated salt solution to obtain desorption isotherm curves. For modeling the desorption isotherm the following GAB (Guggenheim-Anderson-de Boer) equation was used (19).

$$m=\frac{CKm_1a_w}{(1-Ka_w)(1-Ka_w+CKa_w)}$$

in which C and K are constants, and m and m_1 are the equilibrium moisture content and monolayer moisture content, respectively. The parameters of the GAB equation were evaluated by rearranging the equation into a polynomial form then regressing a_w/m against a_w.

$$\frac{a_w}{m}=\frac{K}{m_1}\left(\frac{1}{C}-1\right)a_w^2+\frac{1}{m_1}\left(1-\frac{2}{C}\right)a_w+\frac{1}{m_1KC}$$

Moisture content The vacuum oven method was used to determine the moisture content of the rehmannia samples. Rehmannia samples were placed in pre-dried aluminum dishes in a vacuum oven, with sulfuric acid as a desiccant. The sample was kept at 70°C with a gauge pressure of 85 kPa for 24 hr (17). The samples were then taken out of the oven, cooled in a desiccator and weighed using an electronic balance (MC1 Analytic 210S; Satorius AG, Göttingen, Germany) with a sensitivity of 0.0001 g. The fresh and bone-dried weights were used to calculate the moisture content, which was expressed as g water/g dry solid.

Drying experiments Drying experiments were performed using a temperature controlled drying oven (HB-502 M; Hanback Co., Yongin, Gyeonggi, Korea) at 40, 60, 80, and 100°C and relative humidity of 17.2, 7.0, 2.9, and 1.2%, respectively. Air velocity was kept at 0.45 m/sec. The drier was run without the sample for about 1 hr to set the desired drying conditions before each drying experiment. Then about 60 g (corresponding to 2-3 roots) of each rehmannia sample was uniformly spread in an aluminum pan in a single layer. The pan was hanged over inside the drying chamber and connected to the hook of an electronic balance (Satorius AG) on top of the drying oven using nylon strings. The change in the sample weight was recorded every 5 min during drying with a sensitivity of 0.0001 g. The drying was continued until there was no large variation in the weight change. The sample weight loss during drying was converted into the moisture content on dry basis and expressed as g water/g dry solid.

Analysis of drying data The drying data were analyzed to study the drying behavior of the rehmannia. Generally, a relationship analogous to Newton's law of cooling is often used in drying analysis to describe the falling rate period. The rate of moisture loss is assumed proportional to the moisture remaining to be lost, as follows:

$$dm/dt = -k(m - m_e)$$

and on integration this yields the exponential drying equation, which is known as the Lewis model for thin layer drying of products (20).

$$(m - m_e)/(m_o - m_e) = \exp(-kt)$$

The moisture ratio (MR) and drying rate of the rehmannia samples were calculated using following equations:

$$\mathrm{MR}=\frac{m-m_e}{m_o-m_e}$$

$$\text{Drying rate}=\frac{dm}{dt}=\frac{m_{t+dt}-m_t}{dt}$$

where m, m_o, m_e, are the moisture content at any time, initial moisture content, equilibrium moisture content, and m_t and m_{t+dt} are moisture content at time t and $t+dt$ (g water/g dry solid), respectively, t is drying time (min).

In the falling rate period, diffusion is the dominant physical mechanism governing moisture movement in the sample. The diffusion coefficient (D_{eff}) is usually determined from the slope of a linear line obtained by plotting experimental drying data in terms of ln(MR) versus drying time (21) based on a numerical solution of a partial differential equation, i.e., the Fick's second law of diffusion:

$$\frac{\partial m}{\partial t}=D_{eff}\frac{\partial^2 m}{\partial x^2}$$

where, D_{eff} is the effective diffusion coefficient (m^2/sec) and x is characteristic dimension of the product.

Though the simple exponential relationship has been widely used to explain the drying behavior of various food materials (22), most experimental drying data, especially for most fruits and vegetables, are deviates from the linearity when plotted ln(MR) vs. time (8, 11-14, 23, 24). To overcome the problem in the case of non-linear drying curves, various modified or empirical drying models have been developed. Some of them are as follows:

Page model (12): $MR = \exp(-kt^n)$

Henderson and Pabis' model (13, 25): $MR = a\exp(-kt)$

Logarithmic model (14, 26): $MR = a\exp(-kt) + c$

Two-term model (27): $MR = a\exp(-k_0t) + b\exp(-k_1t)$

Most of these models have been tested to find well fitted to the experimental drying data (8, 12, 14). In this study, the experimental drying data of the rehmannia were tested to fit the logarithmic model, since it is simple to apply and known to fit well to the experimental drying data (14). Parameters (a, k, and c) of the model were evaluated by a non-linear least square regression analysis method based on the Levenberg-Marquardt algorithm using Origin® computer program (OriginLab Corp., Northampton, MA, USA). The goodness of fit was determined using a coefficient of determination (R^2).

Temperature dependency of the drying rate (k) was tested using the Arrhenius equation.

$$k = k_o\exp(E_a/RT)$$

where, k_o is the pre-exponential factor, E_a is the activation energy (J/mol), R is the universal gas constant (8.314 J/mol·K), and T is the absolute temperature (K).

Color measurement The surface color of fresh and dried rehmannia samples were measured in terms of Hunter L (degree of lightness), a (degree of redness), and b (degree of yellowness) values, using a colorimeter (CR-300 Minolta Chroma Meter; Monolta Camera Co., Osaka, Japan). The colorimeter was calibrated against a standard calibration plate of a white surface with L, a, and b values of 98.86, -0.02, and 1.99, respectively. The total color difference (ΔE) was calculated using the following formula with Hunter L, a, and b values obtained.

$$\Delta E = \sqrt{(L_{sample}-L_{standard})^2+(a_{sample}-a_{standrad})^2+(b_{sample}-b_{standrad})^2}$$

where, standard values of L, a, b were used for those of standard white plate. The measurement of color were replicated 5 times after shaking the dried samples and the average and standard deviation values of L, a, b, and ΔE were reported.

Results and Discussion

Desorption isotherm Desorption isotherm curves of the sliced and whole root of rehmannia samples measured at 20°C for a range of water activity between 0.113 to 0.946 are shown in Fig. 1. These curves show biphasic patterns of moisture change against water activity, *i.e.*, the equilibrium moisture content increased very slightly until water activity around 0.75, then increased abruptly. This shape is similar to the Type III isotherms classified by Brunauer *et al.* (28), which is typical for a pure crystalline substance (*e.g.*, glucose). The GAB equation was used to model the desorption isotherms, since the GAB model has been used frequently in food materials and usually provided the best fit (29-32). Table 2 shows the result of parameters of GAB equation for both the sliced and whole root of rehmannia determined at 20°C. The monolayer moisture content of the sliced and whole root of rehmannia was 0.06 and 0.08 g water/g dry soild, respectively. High values of the coefficient of determination (R^2) indicates a good fit of the GAB model to the experimental data of the desorption isotherms.

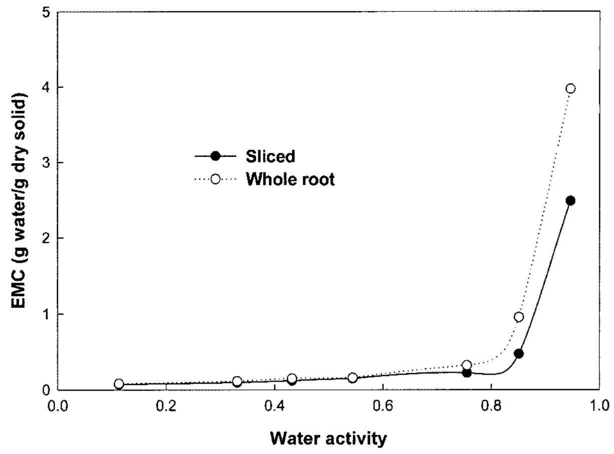

Fig. 1. Moisture desorption isotherms for the sliced and whole root of rehmannia at 20°C.

Drying characteristics The moisture contents of the rehmannia as a function of drying time at different temperatures are presented in Fig. 2 for the sliced and whole root of rehmannia samples. The moisture content of both rehmannia samples decreased exponentially with increase in progress of drying. And the drying time to reach a certain level of moisture content decreased significantly with increase in drying temperature. As expected, the sliced sample required less drying time for dehydration of the rehmannia due to the increased surface area for moisture evaporation. Drying characteristics of the rehmannia samples is more clearly observed in Fig. 3, which shows a relationship between the drying rate vs. moisture content. The effect of increasing the drying temperature on drying rate is evident in both sliced and whole root of samples. As shown clearly in Fig. 3, after an initial short period of settling down (about 1 hr) during which the solid surface conditions come into equilibrium

Table 2. Parameters of the GAB model for desorption isotherm of the rehmannia determined at 20°C

	Sliced	Whole root
m_l	0.06	0.08
C	-758.09	50.31
K	1.02	1.05
R^2	0.99	0.98

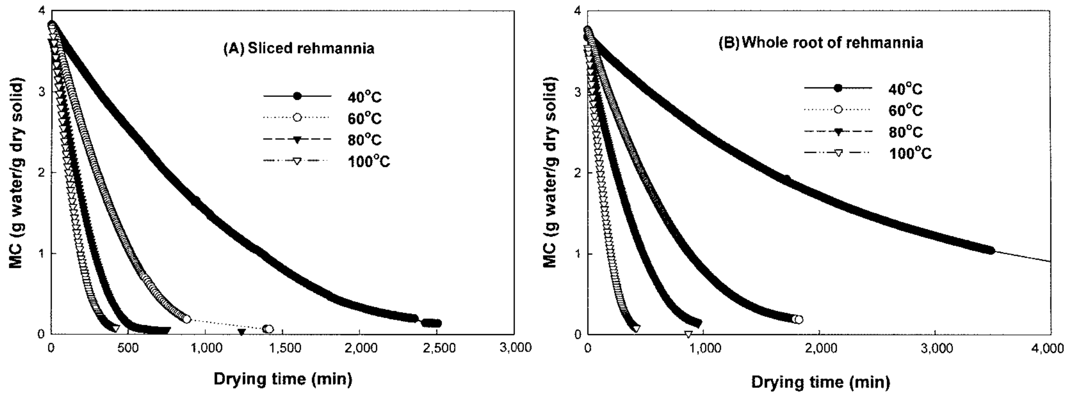

Fig. 2. Drying curves for the sliced and whole root of rehmannia.

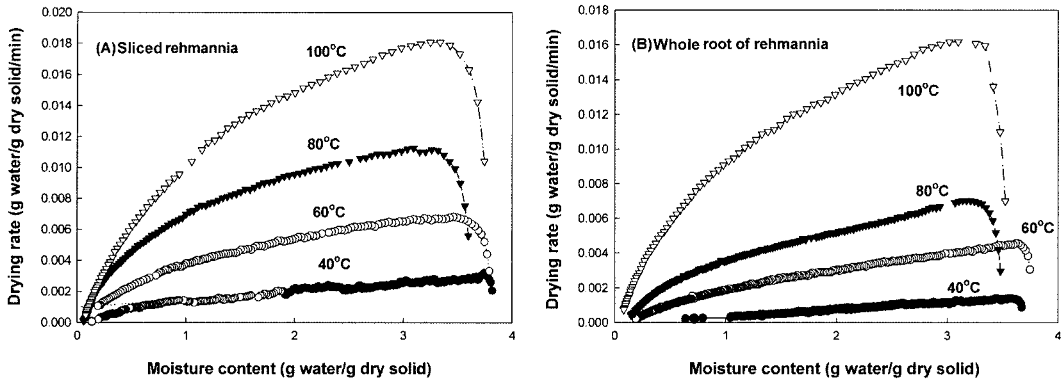

Fig. 3. Drying rate curves for the sliced and whole root of rehmannia.

with the drying air, the drying rate reached a maximum value then decreased afterward. The periods of drying that was observed were mostly falling rate periods in both rehmannia samples. The period of constant drying rate, which is frequently observed in the drying of fruits, was not observed at all. This indicates that diffusion-controlled process is dominant, *i.e.*, the rate of moisture removal is controlled by diffusion of moisture from inside to the surface of the rehmannia (22). Similar drying behavior was also observed in the drying of various other fruits and vegetables such as red pepper (4, 5), garlic (8), okra (12), fig (13), mulberry (14), grapes (33), and apricots (34), etc.

Dehydration kinetic parameters For the analysis of experimental drying data for the rehmannia samples using thin layer drying model, the equilibrium moisture contents (EMC) of the rehmannia samples were determined first. The EMCs of the sliced and whole root of rehmannia as a function of relative humidity at the drying temperatures were calculated using the GAB equation with the parameters evaluated at 20°C (4, 6) and the results are shown in Table 3. The EMCs were ranged from 0.069 to 0.078 g water/g dry solid depending on the drying temperature, and they were coincided with the monolayer moisture content of the rehmannia evaluated from the desorption isotherms using the GAB model.

Initial trial for using the Lewis model to analyze the experimental drying data of the rehmannia samples was failed since non-linear lines were observed when the experimental moisture ratio (MR) was plotted against drying time (t). Therefore the drying data were fitted to the logarithmic model to evaluate the parameters for the model and the values are tabulated in Table 4. The analysis yielded high values of R^2, implying a good fitness of the logarithmic model to the experimental data. The drying times for the rehmannia samples to reach the equilibrium moisture content (t_{EMC}) at each drying temperature were evaluated using the logarithmic model. The results are shown in Table 3. The t_{EMC} values for the sliced and whole root of rehmannia indicate that they decreased exponentially with increase in drying time. This result also shows that the drying time of the sliced sample was shorter than that of the whole root of rehmannia by 3.1 to 1.1 times depending on drying temperature. The difference in drying time between the sliced and whole root of

Table 3. Estimated equilibrium moisture content (EMC) of the rehmannia and drying time to reach EMC (t_{EMC}) at different drying temperature

		EMC (g water/g dry solid)		t_{EMC} (min)	
T (°C)	a_w	Sliced	Whole root	Sliced	Whole root
40	0.172	0.078	0.071	3161.5	9831.3
60	0.070	0.070	0.069	1318.7	2299.4
80	0.029	0.069	0.069	657.7	1191.4
100	0.012	0.073	0.070	432.5	480.8

Table 4. Parameters of the logarithmic model for drying kinetics of sliced and whole root of the rehmannia

	Sliced				Whole root			
	40°C	60°C	80°C	100°C	40°C	60°C	80°C	100°C
a	4.6619	5.0166	4.4978	4.5056	3.4200	4.1616	4.1313	4.3483
k	-0.00071	-0.000168	-0.00353	-0.00577	-0.00043	-0.00130	-0.00211	-0.00503
c	-0.7841	-1.0719	-0.6354	-0.4644	-0.2830	-0.2983	-0.5052	-0.5750
R^2	0.9990	0.9987	0.9951	0.9954	0.9999	0.9988	0.9985	0.9967

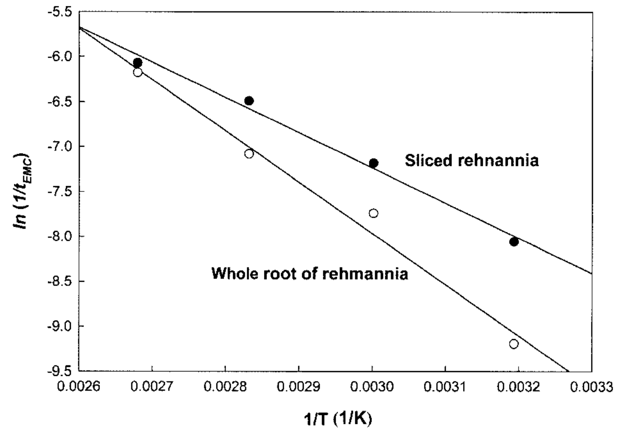

Fig. 4. Temperature dependency of the drying time to reach the equilibrium moisture content (t_{EMC}) for the sliced and whole root of rehmannia.

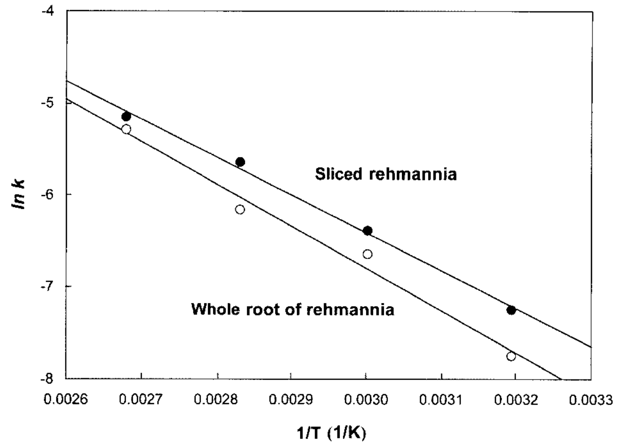

Fig. 5. Temperature dependency of the drying rate constant (k) for the sliced and whole root of rehmannia.

rehmannia decreased with increase in drying temperature. The decrease in drying time of the sliced sample may be due to increase in surface area for moisture removal and also due to the less resistance against moisture evaporation at the surface of exposed endocarp. When the products are dehydrated whole, the pericarp has the effect of an external resistance and as a consequence of increased drying time. In the case of sliced rehmannia the inner face was exposed to the air stream, favoring moisture removal during the drying process. Sanjuán *et al.* (4) also observed the time needed to dehydrate whole peppers was much longer (nearly 10 times more) than that for shredded peppers. The less effect of reducing drying time by slicing for the rehmannia than pepper may be attributable to the less moisture vapor resistant pericarp of the rehmannia.

Interestingly, the inverse of time to reach the equilibrium moisture content ($1/t_{EMC}$) indicated linear temperature dependency described by the Arrhenius type equation as shown in Fig. 4 with following relationship:

$ln\ (1/t_{EMC}) = -3916.1\ (1/T) + 4.5108$ for sliced rehmannia
$ln\ (1/t_{EMC}) = -5669.4\ (1/T) + 9.0397$ for whole root of rehmannia

For these relationships, the activation energy and frequency factor were determined and tabulated in Table 5. This result suggests that this relationship can be conveniently used to determine optimum drying time at different drying conditions.

In addition, the drying constant k (1/min) determined using the logarithmic model (Table 4) also found to be followed the Arrhenius equation as shown in Fig. 5. The kinetic parameters for the drying of rehmannia samples were determined and also shown in Table 5. The activation energy values for the sliced and whole root of rehmannia are 34.27 and 38.26 kJ/mol, respectively. This method for determination of Arrhenius kinetic parameters using the drying constant (k) is much simpler to apply than using diffusion coefficient (D_{eff}) for drying. This method is not only simple to apply but also no need for unrealistic assumptions, such as the system is isotropic and the initial moisture content is uniform and the drying surface reaches its equilibrium values instantaneously and there is no shrinkage, needed in determination of D_{eff} values. The activation energy determined using k values are very similar to those obtained for D_{eff} values (13). For example, Babalisi and Belessiotis (13) reported activation energy values evaluated with D_{eff} values for drying fig as 32.27, 30.81, 45.81, and 48.47 kJ/mol determined at different air

Table 5. Arrhenius kinetic parameters determined using time to reach equilibrium moisture content (t_{EMC}) and drying constant (k) of the rehmannia

		Activation energy (kJ/mol)	Frequency factor (1/min)	R^2
$1/t_{EMC}$	Sliced	32.56	0.91×10^2	0.9899
	Whole root	47.14	84.31×10^2	0.9842
k	Sliced	34.27	3.86×10^2	0.9954
	Whole root	38.26	11.02×10^2	0.9842

Table 6. Effect of drying temperature on surface color attribute of sliced and whole root of the rehmannia[1)]

	L	*a*	*b*	*ΔE*
Sliced				
Fresh	71.64 ± 0.78^{h}	3.71 ± 0.11^{b}	32.51 ± 1.15^{e}	39.77 ± 1.39^{a}
40°C	44.55 ± 1.81^{e}	5.58 ± 0.46^{d}	24.77 ± 0.21^{d}	57.33 ± 1.68^{c}
60°C	46.10 ± 0.85^{ef}	4.46 ± 0.30^{c}	24.83 ± 0.15^{d}	55.84 ± 0.74^{c}
80°C	41.64 ± 0.80^{d}	4.26 ± 0.44^{bc}	19.48 ± 0.43^{c}	58.09 ± 0.82^{c}
100°C	37.08 ± 2.07^{c}	4.25 ± 0.37^{bc}	20.27 ± 1.26^{c}	62.67 ± 2.00^{d}
Whole root				
Fresh	54.76 ± 0.44^{g}	14.36 ± 0.11^{e}	36.27 ± 0.74^{f}	56.16 ± 0.46^{c}
40°C	47.65 ± 0.78^{f}	5.38 ± 0.35^{d}	15.69 ± 0.29^{b}	51.37 ± 0.78^{b}
60°C	28.53 ± 0.75^{b}	2.56 ± 0.40^{a}	10.10 ± 0.20^{a}	68.86 ± 0.77^{e}
80°C	23.67 ± 2.11^{a}	2.62 ± 0.09^{a}	10.58 ± 0.30^{a}	73.74 ± 2.12^{f}
100°C	22.68 ± 1.43^{a}	2.66 ± 0.31^{a}	10.19 ± 0.28^{a}	74.68 ± 1.40^{f}

[1)] Means of 5 replicates±SD; Any two means in the same column followed by the same letter are not significantly different ($p>0.05$) by Duncan's multiple range test.

velocity of 0.5, 1, 2, and 2 m/sec, respectively. Activation energy values evaluated with *k* values determined by the Henderson and Pabis' model using the same experimental drying data were 36.4, 30.3, 45.3, and 53.2 kJ/mol, respectively. Many researchers also used the values of the drying constant (*k*) obtained using drying models to determine kinetic parameters using Arrhenius type equation (35-39). The kinetic parameters therefore can be considered as a pseudo-diffusivity for drying of food materials.

Temperature effect on surface color Effect of drying temperature on surface color of rehmannia determined as Hunter color values are shown in Table 6. Initially, sliced sample shows higher *L*-values and lower *a*-values than those of whole root of rehmannia. This is mainly because reddish-brown color of rehmannia skin. As drying progress surface color of rehmannia turned dark due to browning reaction. It was found that with an increase in the drying temperature, the color became darker implying that more browning of rehmannia occurred. It has also reported in the literature that browning increase with an increase in the drying temperature and time (40). This result suggests that surface color values can be properly used as an index for suitable drying of rehmannia.

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