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Evaluation of semi-empirical and non-linear drying models by Bayesian inference

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ABSTRACT. Current analysis investigates thin layer drying experiments on passion fruit seeds. Drying characteristics of passion fruit seeds were examined using ambient air for temperature range between 40 and 65ºC and air flow velocity range between 0.6 and 1.2 m s^{-1}. The semi-empirical models were then fitted to the drying data by Bayesian inference, based on the ratios of the difference between the initial and final moisture contents and equilibrium moisture content. The effective diffusivity varied between 1.70 x 10^{-10} and 4.68 x 10^{-10} m^2 s^{-1} with temperature and air flow rate increase. Temperature dependence on the diffusivity coefficient was described by an Arrhenius-type relationship. The activation energy for moisture diffusion was 24.36, 35.24 and 13.86 kJ moL^{-1} for drying air speeds respectively equal to 0.6, 0.9 and 1.2 m s^{-1}.

Keywords: passion fruit seeds, drying, diffusivity, Lewis model.

Avaliação de modelos semi-empíricos para secagam usando inferência Bayesiana

RESUMO. Nesse trabalho, foram realizados experimentos de secagem da semente do maracujá amarelo em camada delgada. Foram examinadas as características da secagem das sementes de maracujá, usando ar ambiente na faixa de temperatura de 40-65ºC e velocidade do ar de secagem na faixa de 0,6-1,2 m s^{-1}. Os modelos semiempíricos foram ajustados aos dados por inferência Bayesiana, baseados nas razões de diferenças de umidades inicial e final e no equilíbrio de umidade. A difusividade efetiva variou de 1.70 x 10^{-10} a 4.68 x 10^{-10} m^2 s^{-1} com o aumento de temperatura. A difusividade apresentou dependência do tipo Arrhenius com a temperatura. As energias de ativação encontradas foram 24.36, 35.24 e 13.86 kJ moL^{-1} para as velocidades de ar de secagem iguais a 0,6, 0,9 e 1,2 m s^{-1}, respectivamente.

Palavras-chave: semente de maracujá, secagem, difusividade, modelo de Lewis.

Introduction

Passion fruit (*Passiflora edulis*), a Brazilian native plant, is a popular tropical fruit known worldwide. In the juice industry, the passionfruit produces thousands of tons of seeds as agricultural byproducts during juice extraction. The seeds, which contain large amounts of oil, are generally discarded. Since they total several metric tons, the byproduct is an asset of great economical, scientific and technological interest.

The basic objective in drying food products is the removal of water from the solids up to a certain level at which microbial spoilage and deterioration of chemical reactions are greatly minimized (KRODIDA et al. 2003). Convective air-drying is the most frequently used dehydration method in the food and chemical industry, although a considerable amount of data has been reported in the literature on thin-layer drying of different agricultural products such as eggplant (ERTEKIN; YALDIZ, 2004), green bean (ROSSELLÓ et al., 1997; SENADEERA et al., 2003), green peas (SIMAL et al., 1996), okra (ADOM et al., 1997; DOYMAZ, 2005; GOGUS; MASKAN, 1999), red pepper and red chili (DOYMAZ; PALA, 2002; GUPTA et al. 2002), sweet potato (DIAMANTE; MUNRO, 1993); black tea (PANCHARIYA et al., 2002), carrots (DOYMAS, 2004a), white mulberry (DOYMAZ, 2004b) and kiwi fruit (SIMAL et al., 2005).

The choice of an appropriate statistical method for modeling non-linear curves used for the drying processes is of paramount importance in obtaining results. In fact, Bayesian inference is a good option as opposed to frequentist methods (least squares), because tab is based on iterative processes, or alternatively, means are used to linearize the model by logarithmic transformation (SILVA et al., 2005; ROSSI, 2011).

Current study reports the effect of drying temperature and air flow rate on the drying characteristics of passion fruit seeds to evaluate a suitable drying model by Bayesian inference, in order to describe the drying process and compute effective moisture diffusivity and activation energy of passion fruit seeds while drying.

Mathematical modeling

In current study, the falling rate period was not observed in any of the experiments. In most studies on drying, diffusion is generally accepted to be the main mechanism during the transport of moisture to the surface to be evaporated. The solution of Fick's equation is presented below, with the assumptions of moisture migration by diffusion, negligible shrinkage, constant diffusion coefficients and temperature and for a sphere (CRANK, 1975; DOYMAZ, 2004a; PALA et al., 1996):

$$MR = \frac{M - M_e}{M_o - M_e} = \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} \exp\left(- \frac{n^2 \pi^2 D_{eff} t}{r^2} \right) \quad (1)$$

where:

M, M_o and M_e are content, initial and equilibrium moisture content, respectively; r is the radius; t is the time; MR is the moisture ratio; D_{eff} is the effective diffusivity.

The semi-theoretical models are generally derived by simplifying general series solutions of Fick's second law or the modification of simplified models and valid within the temperature, relative humidity, air flow velocity and moisture content range for which they were developed (FORTES; OKOS, 1981). These models require a shorter time when compared to theoretical thin-layer models and do not need assumptions of geometry of a typical food, its mass diffusivity and conductivity (PARRY, 1985). There are several semi-theoretical thin-layer drying models, although in current study Lewis's model has been employed: Lewis's model with two parameters and Lewis's model with three parameters. Models are shown in Table 1.

Table 1. Thin-layer drying models.

	Model	Equation
M_1	Lewis	$MR = \exp(-k_1 t)$
M_2	Lewis with two parameters	$MR = a_2 \exp(-k_2 t)$
M_3	Lewis with three parameters	$MR = a_3 \exp(-k_3 t) + b_3$

Where k_1, k_2 and k_3 are diffusivity and a_2, a_3 and b_3 are model constants.

Bayesian inference

In general, the drying seeds have been carried out from a frequentist approach, adjusting nonlinear models which aim at synthesizing pieces of information into parameter estimates to be interpreted. According to Oliveira et al. (2012a and b; 2013), estimation is based on iterative processes such as those by Gauss-Newton, DUD and Marquardt algorithm, due to the nonlinearity of the variables. These procedures minimize the sum of residue squares. However, when individual adjustments are considered, i.e. adjustments for many experimental units of mathematically complex models or few possible longitudinal observations are extant, interactive methods frequently provide negative estimates for parameters which may cause the formation of atypical curves.

Furthermore, when dealing with comparisons of curves derived from different treatments, the distribution of nonlinear model parameter estimators do not usually follow Gaussian distribution. Therefore, the process to formulate statistical tests becomes complex once presuppositions related to the asymptotic theory are not attended (OLIVEIRA et al., 2012a and b). Bayesian inference, involving the adjustment of linear and nonlinear regression models, has been successfully used in recent years since it reduced the number of biased estimations even when little information was used (OLIVEIRA et al., 2012a and b; 2013).

Material and methods

The laboratory dryer

The experimental apparatus used for convective drying of passionfruit seeds has been described by Fiorentin et al. (2010).

Drying experiments

A local factory provided the passionfruit seeds which were washed to remove the juice remaining on them. The cleaned seeds were then pre-dried under sunlight at day temperature, about 32 ± 2°C, for 5 hours. The seeds were later dried on thin-layer.

Experiments were performed to determine the effect of process variables on thin-layer drying characteristics of passionfruit seeds. Four temperatures (40, 50, 60, and 65°C) were studied, with constant air flow rate at three points (0.6, 0.9, and 1.2 m s^{-1}), at constant air temperature. Twenty-four drying runs were performed in a systematic way, constituting two replicates. An anemometer with a reading accuracy of ± 0.05 m s^{-1} measured air velocity, with measurement location being 60 cm above the plenum of the test chamber. The temperature of air entering the plenum chamber was measured by thermocouple.

The initial moisture content of passionfruit seeds was measured according to Brazilian rules for seeds (BRASIL, 1992). The 500 g sample was uniformly spread in a basket in a single layer after the desired drying conditions had stabilized. Water losses were measured by weighing the basket and its content at 3-minute intervals until constant weight. These tests were performed in duplicate and the averages were reported and used at the simulation modeling.

Mathematical modeling by Bayesian inference

For the first stage it was assumed that *Y:MR* has normal distribution truncated within the range [0,1], i.e:

$$Y_i \sim Normal\left(\mu(t_i), \sigma_e^{\,2}\right)_{[0,1]} \tag{2}$$

$\sigma_e^2 = \tau_e^{-1}$ OpenBugs parameterization, $E\left(Y\right) = \mu(t) =$ proposed model. The models are shown in Table 1.

For all model parameters the *prior* was considered non-informative Gamma distribution (default OpenBugs), for strictly positive quantities, i.e.:

$$parameters \sim Gamma\left(10^{-3}, 10^{-3}\right)$$

The posterior distributions of the parameters were obtained by BRugs on R program (R DEVELOPMENT CORE TEAM, 2012). One million samples were obtained by Monte Carlo Markov Chain (MCMC), where 100,000 were discarded ("burn-in samples") to eliminate the effect of the initial values. The final samples were taken with steps of 100 which contained 9,000 values. The convergence chains were verified by Convergence Diagnosis and Output Analysis - CODA program - (BEST et al., 1995) by Geweke (1992) and Heidelberger and Welch (1983) criteria.

Deviance Information Criterion (DIC) may be used as a comparison, as well as the selection of (co)variables in models. Speigelhalter et al. (2002) suggested the use of difference criterion module between DIC values of two models, A and B, analyzed. This criterion is shown in Equation 3.

$$D = \left|DIC_A - DIC_B\right| \tag{3}$$

If D < 5, it may be concluded that there was no significant difference; if $5 \le D \le 10$, it may be concluded that there was significant difference; if D > 10, it may be concluded that there was high significant difference.

Results and discussion

Influence of process parameters

Table 2 shows the initial and final moisture conditions of passionfruit seeds.

Table 2. Initial and final moisture conditions of passionfruit seeds in different conditions, on dry base.

Velocity ($m\ s^{-1}$)	Temperature (°C)	Initial moisture (%)	Final moisture (%)
0.6	40	11.1	8.51
	50	12.9	11.0
	60	14.0	12.6
	65	14.0	8.46
0.9	40	16.4	6.34
	50	12.3	7.39
	60	10.1	6.95
	65	10.1	7.11
1.2	40	8.20	5.11
	50	8.20	5.73
	60	8.00	5.13
	65	8.00	5.41

Drying kinetics of the curves do not present constant drying period, or rather, the time spent during the drying process was only for removing internal moisture. The constant rate was reported by various authors (DOYMAZ 2005; 2004a and b). When there is an absence of constant period, the diffusion is the physical mechanism that governs the drying process. Mandamba et al. (1996); Gouveia et al. (2003); Doymaz (2004a and b; 2005); Ertekin and Yaldiz (2004); Simal et al. (2005) also noted this behavior.

Increase in the drying temperature caused an important increase in the drying rate; consequently, drying time is decreased. The effect of air flow rate was more sensitive when compared to the air flow rate of 0.6 and 0.9 m s^{-1}. In these conditions, the increase of air flow rate caused a decrease on drying time.

For a speed of drying air at 0.6 m s^{-1}, the time necessary for the establishment of a constant mass condition in the system was approximately 64% higher for the temperatures 40, 50, and 60°C, when they were compared to a temperature of 65°C. In the case of speed at 0.9 m s^{-1}, the time required for the system tray/seed to reach constant mass condition was about 73% higher for the temperature 40°C when the latter is compared to the temperatures 50, 60, and 65°C. At 1.2 m s^{-1}, the time necessary for reaching the constant mass condition at the system decreased 44% when the temperature of 40°C was compared to the temperature 65°C.

In general, the increase on the air speed results in a drying rate increase, which is represented by the declivity of the line tangent to the drying curve. If the temperature is kept constant and the drying air speed varies between 0.6 and 1.2 m s^{-1}, there is a reduction of 39% on the drying time. A similar behavior may be observed for the temperatures 50, 60 and 65°C, with reductions approximately equal to 64, 50 and 22%, respectively.

These results are only valid for drying in thin layer. They cannot be applied to drying in thicker layers, in which other variables may have an influence on the time the seed is dried.

Evaluation of the models

So that moisture content as a function of drying time could be determined, the models of Table 1 were fitted according to Bayesian inference. The Bayesian estimations (*posterior*) to the model parameters to drying (mean, standard deviation – SD- and DIC) are given in Tables 3 to 5 for different conditions.

The comparison of the DICs obtained, listed in Tables from 3 to 5, at the diverse experimental conditions, by means of Equation 3, showed there was no significant difference among the models evaluated. As Lewis's model presented only one parameter to be adjusted, it should be used for describing the drying kinetics of passionfruit seed in a thin layer.

It may be observed in Tables 3 to 5 that the mean obtained was close to the median for all the parameters of the models studied. It showed a symmetrical distribution. It may also be verified that zero did not belong to the credibility intervals of the parameters of the models studied, in all the conditions. This means that all parameters studied were statically not null.

When Lewis's model (M_1, M_2 and M_3) and the three air drying speeds are taken into account, it may be verified that the drying coefficient (k_1, k_2 and k_3) varied with the variation of absolute temperature (in Kelvin). The variation from 40 to 50°C was a significant temperature increase, whereas the increases from 50 to 60, and to 65°C were not so significant. Henderson and Pabis (1961) demonstrated that the drying coefficient k is not constant, but dependent on the drying air temperature and on the mechanisms of water diffusion or water vapor inside the seeds.

Figure 1 shows the drying coefficient variation (*k's*) with variations of absolute temperature (in Kelvin) for the speeds of drying air under analysis. The adjustment equations and R^2 (determination coefficients) are presented in Table 6. In their experiments on drying kiwi fruit, Simal et al. (2005) observed that variation of *k's* with temperature exhibited a linear trend.

Table 3. Bayesian estimations model parameters on drying of passionfruit seeds at 0.6 m s^{-1} at different temperatures.

Temperature (°C)		40			50			60			65		
Model	Parameter	Mean	DIC	SD	Mean	DIC	SD	Mean	DIC	SD	Mean	DIC	SD
M_1	k_1	0.000641	-57.67	5.09×10^{-5}	0.001525	-33.20	2.04×10^{-4}	0.00144	-29.40	2.87×10^{-4}	0.001476	-23.52	1.31×10^{-3}
M_2	a_2	0.862800	-54.96	4.62×10^{-2}	0.896200	-33.04	7.79×10^{-2}	0.871400	-29.44	8.73×10^{-2}	0.86980	-23.70	9.82×10^{-2}
	k_2	0.000517		5.47×10^{-5}	0.001319		2.31×10^{-4}	0.001194		2.48×10^{-4}	0.00120		2.90×10^{-4}
M_3	a_3	0.859300	-56.62	3.43×10^{-2}	0.882700	-35.11	6.59×10^{-2}	0.862200	-32.67	6.90×10^{-2}	0.847100	-27.57	6.95×10^{-2}
	b_3	0.018130		1.91×10^{-2}	0.031710		2.83×10^{-2}	0.045670		3.54×10^{-2}	0.095110		4.27×10^{-2}
	k_3	0.000565		6.31×10^{-5}	0.001545		3.64×10^{-4}	0.001528		3.94×10^{-4}	0.0019880		7.87×10^{-4}

Table 4. Bayesian estimations model parameters on drying of passionfruit seeds at 0.29 m s^{-1} at different temperatures.

Temperature (°C)		40			50			60			65		
Model	Parameter	Mean	DIC	SD	Mean	DIC	SD	Mean	DIC	SD	Mean	DIC	SD
M_1	k_1	0.0009819	-32.5	1.18×10^{-4}	0.001771	-30.68	2.08×10^{-4}	0.00199	-31.96	2.33×10^{-4}	0.00184	-31.35	2.15×10^{-4}
M_2	a_2	0.8523000	-34.81	6.49×10^{-2}	0.926700	-29.91	6.20×10^{-2}	0.941400	-30.26	6.66×10^{-2}	0.935900	-29.72	6.49×10^{-2}
	k_2	0.0007791		1.24×10^{-4}	0.001611		2.17×10^{-4}	0.001857		2.75×10^{-4}	0.001705		2.52×10^{-4}
M_3	a_3	0.847000	-37.2	6.27×10^{-2}	0.925000	-30.68	6.22×10^{-2}	0.929000	-31.63	6.07×10^{-2}	0.928300	-33.36	5.69×10^{-2}
	b_3	0.0394300		3.45×10^{-2}	0.021290		2.62×10^{-2}	0.021880		2.46×10^{-2}	0.036680		2.90×10^{-2}
	k_3	0.0009542		2.13×10^{-4}	0.001799		3.23×10^{-4}	0.002049		4.37×10^{-4}	0.002028		3.52×10^{-4}

Table 5. Bayesian estimations model parameters on drying of passionfruit seeds at 1.29 m s^{-1} at different temperatures.

Temperature (°C)		40			50			60			65		
Model	Parameter	Mean	DIC	SD	Mean	DIC	SD	Mean	DIC	SD	Mean	DIC	SD
M_1	k_1	0.001155	-36.51	9.29×10^{-5}	0.001553	-33.02	1.49×10^{-4}	0.001498	-34.88	1.11×10^{-4}	0.001747	-27.22	2.11×10^{-4}
M_2	a_2	0.920600	-37.16	4.84×10^{-2}	0.930800		5.82×10^{-4}	0.943000	-33.83	5.03×10^{-2}	0.932400	-26.10	6.57×10^{-2}
	k_2	0.001039		1.05×10^{-4}	0.001430	-32.13	1.78×10^{-4}	0.001405		1.32×10^{-4}	0.001616		2.43×10^{-4}
M_3	a_3	0.92080	-37.19	4.70×10^{-2}	0.925700		5.03×10^{-2}	0.933700	-34.35	4.75×10^{-2}	0.925300	-27.26	5.66×10^{-2}
	b_3	0.01330		2.08×10^{-2}	0.015160		2.27×10^{-2}	0.009098		1.66×10^{-2}	0.020580		2.92×10^{-2}
	k_3	0.00110		1.43×10^{-4}	0.001523	-32.76	2.30×10^{-4}	0.001453		1.62×10^{-4}	0.001749		4.06×10^{-4}

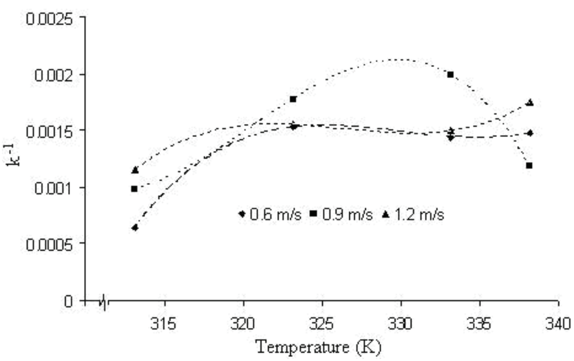

Figure 1. Dependence on Lewis's parameter (k_1, k_2 and k_3) at absolute temperature.

Table 6. Adjustment equations of k_1, k_2 and k_3 parameters.

Velocity ($m\ s^{-1}$)	Adjustment equation	R^2
0.6	$k_1 = 2\times10^{-7}T^3 - 0.0002T^2 + 0.0077T - 8.4623$	0.999
0.9	$k_2 = -4\times10^{-7}T^3 + 0.0004T^2 - 0.1153T + 12.305$	0.999
1.2	$k_3 = 2\times10^{-7}T^3 - 0.0002T^2 + 0.0076T - 8.2664$	0.999

Evaluation of moisture diffusivity and action energy

Finally, effective diffusivity coefficient was determined to establish diffusion model. The solution of Fick's second law (Equation 1) out of spheres was shown. According to several authors (NUH, BRINKWORTH, 1997; PALA et al, 1996; RIVA; PERI, 1986; DOYMAZ, 2005) Equation (1) assumes that the effective diffusivity $\left(D_{eff}\right)$ is constant and shrinkage of the samples is negligible. For long drying time (setting n = 1), Equation (1) could be further simplified to a straight-line equation as:

$$\ln(MR) = \ln\left(\frac{6}{\pi^2}\right) - \left(\frac{\pi^2 D_{eff}}{r^2}t\right) \quad (4)$$

The effective diffusivity was calculated by Equation (4) with slopes derived from linear regression of ln(MR) against time data shown in Figure 2. Table 7 shows rates of D_{eff} for different temperatures and air flow rate.

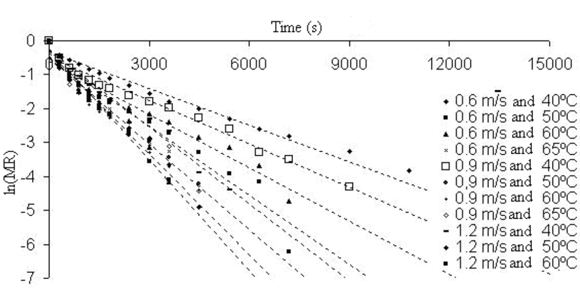

Figure 2. Experimental and predicted logarithmic moisture ratio at different drying time conditions.

Table 7. Effective diffusivity at different conditions.

Velocity ($m\ s^{-1}$)	Temperature (ºC)	D_{eff} ($m\ s^{-1}$)
0.6	40	1.70×10^{-10}
	50	2.55×10^{-10}
	60	2.12×10^{-10}
	65	3.40×10^{-10}
0.9	40	1.70×10^{-10}
	50	3.40×10^{-10}
	60	3.83×10^{-10}
	65	3.40×10^{-10}
1.2	40	3.41×10^{-10}
	50	4.26×10^{-10}
	60	4.68×10^{-10}
	65	4.26×10^{-10}

Increase of temperature means an increase on effective diffusivity $\left(D_{eff}\right)$ at constant air velocity. Consequently, there is a reduction of internal resistance of drying with temperature increase. The figures for diffusivity coefficient proposed by different authors and reported by Pavón-Melendez et al. (2002) varied between 2.2 x 10^{-10} and 9.4 x 10^{-10} $m^2\ s^{-1}$ for different fruits and vegetables such as grapes, potatoes, apples and carrots. Similar behaviors were reported by Panchariya et al. (2002) with regard to black tea; by Doymaz (2004a) on carrots; by Doymaz (2004b) on white mulberry; by Doymaz (2005) on okra and by Simal et al. (2005) on kiwi fruit.

According to several authors, temperature dependence on diffusivity coefficient follows the Arrhenius equation (Equation 5), where E_a is the activation energy (kJ moL^{-1}); D_0 is the pre-exponential factor ($m^2\ s^{-1}$); T is the absolute temperature (K); R is the gas constant (kJ moL K^{-1}). To obtain the temperature influence on effective diffusivity, the rates of $\ln\left(D_{eff}\right)$ are plotted versus $1/T$, as shown in Figure 5.

$$D_{eff} = D_0 \exp\left(-\frac{E_a}{RT}\right) \quad (5)$$

So that temperature influence on effective diffusivity could be evaluated, $\ln\left(D_{eff}\right)$ rates were plotted versus the reciprocal of absolute temperature, as shown in Figure 3. When obtaining the curves presented in Figure 3, the experimental points for speeds 0.9 and 1.2 m s^{-1} referring to 65ºC were neglected because they did not present Arrhenius-type dependence relationship.

The rates from the lines in Figure 3 and in linearized Equation 5 were used to determine the rates of activation energy at distinct conditions of drying air speed. Rates obtained were 24.36, 35.24

and 13.86 kJ moL^{-1} for drying air speeds equal to 0.6, 0.9 and 1.2 m s^{-1}, respectively. Gupta et al. (2002) reported activation energy of 41.95 kJ moL^{-1} for chili peppers; Simal et al. (1996) found activation energy of 28.40 kJ moL^{-1}, whereas Doymaz (2005) reported an activation energy of 51.26 kJ moL^{-1}.

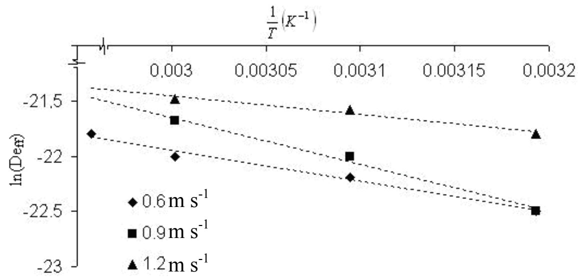

Figure 3. Effect of temperature on moisture diffusivity in passionfruit seeds.

Conclusion

The effect of temperature and air flow rate on the drying of passion fruit seeds was analyzed. The drying process took place only in the falling rate period for passionfruit seeds. Semi-theoretical Lewis, Lewis with two parameters and Lewis with three parameters models were fitted according to Bayesian inference. Although all of them may be used to describe drying behavior, Lewis's model was employed to investigate the dependence relationship of *k* parameter with temperature. The values of effective diffusivity calculated ranged between 1.70 x 10^{-10} and 4.68 x 10^{-10} m^2 s^{-1}. In fact, the effective diffusivity increases as temperature increases. Temperature dependence on diffusivity coefficients was described by Arrhenius relationship and activation energy was found as 24.36, 35.24, and 13.86 kJ moL^{-1} for drying air speeds equal to 0.6, 0.9, and 1.2 m s^{-1}, respectively.

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