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Adjustment of non-linear models for drying in thin layer by bayesian inference

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ABSTRACT. Thin layer air-drying experiments on passion fruit seeds were conducted and the effects of temperature and air-drying speed were investigated in a pilot air-dryer. Drying characteristics of passion fruit seeds were examined by heated ambient air at 40 and 60ºC and air-flow speed at 0.4 and 0.6 m s^{-1}. Analysis of variance showed that the main effects were statistically significant for temperature ($p < 5\%$) and for the interaction between air-speed and temperature ($p < 5\%$). Diffusivity coefficient dependence with temperature was described by an Arrhenius-type equation. Diffusivity coefficient rates were 1.19×10^{-10} m^2 s^{-1}, at 0.9 m s^{-1} and 40ºC, to 1.55×10^{-10} m^2 s^{-1} at 0.6 m s^{-1} and 60ºC. Activation energies were 13.3 and 14.2 kJ mol^{-1} for speeds 0.6 and 0.9 m s^{-1}, respectively.

Keywords: passiflora, drying characteristics, Bayesian inference.

Ajuste de modelos não lineares de secagem em camada delgada por inferência bayesiana

RESUMO. Neste estudo, foi realizada a secagem da semente de maracujá em camada delgada. O efeito da temperatura e da velocidade do ar de secagem foi avaliado em ensaios de secagem numa planta piloto. As características de secagem foram examinadas com ar ambiente aquecido nas temperaturas de 40 e 60ºC e velocidades de 0,4 e 0,6 m s^{-1}. A análise de variância mostrou que os efeitos principais são significativos para a temperatura e para interação temperatura e velocidade do ar de secagem. A dependência do coeficiente de difusividade com a temperatura foi descrito pela equação de Arrhenius. Os valores dos coeficientes de difusividade foram $1,19 \times 10^{-10}$ m^2 s^{-1}, a 0,9 m s^{-1} e 40ºC, e $1,55 \times 10^{-10}$ m^2 s^{-1} a 0,6 m s^{-1} e 60ºC. As energias de ativação encontradas foram de 13,3 e 14,2 kJ mol^{-1} na condição de velocidade de 0,6 e 0,9 m s^{-1}, respectivamente.

Palavras-chave: maracujá, secagem, inferência Bayesiana.

Introduction

The passion fruit (*Passiflora edulis*), native to Brazil albeit a popular tropical fruit worldwide, is usually used for juice production and works best as a flavor in many delicacies. The passion fruit produces a vast number of seeds as agricultural byproducts during juice industrial extraction. Passion fruit seeds containing large amounts of fiber and oil are generally discarded after being crushed.

The drying of agricultural products has always provided a significant income contribution to agricultural societies (BABILIS et al., 2006).

Drying is a widely used industrial preservation method in which the water activity of food is decreased to minimize biochemical reactions of degradation (DOYMAZ; PALA, 2003). Conventional air-drying is the most frequently used dehydration operation in the food and chemical industry (BABILIS; BELESSIOTIS, 2004) because it provides uniformity and hygiene conditions for food industrial processes.

Several studies have been recently published on the drying characteristics of various materials: eggplant (ERTEKIN; YALDIZ, 2004), green bean (ROSSELLÓ et al., 1997; SENADEERA et al., 2003), green peas (SIMAL et al., 1996), okra (ADOM et al., 1996; 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 (DOYMAZ, 2004a), white mulberry (DOYMAZ, 2004b), kiwi (SIMAL et al., 2005), pear (KARATHANOS; BELESSIOTIS, 1999), corn (FORTES; OKOS, 1981).

Theory

Mathematical modeling

Drying process simulation models are used for designing new or improving existing drying systems or even for the control of the drying process (BABILIS et al., 2006). Depending on the applied equations, models may be classified as theoretical, semi-empirical and empirical. Theoretical models take into account only internal resistance to moisture transfer, while semi-empirical and empirical models only consider external resistance to moisture transfer resistance between product and air (PARRY, 1985).

There are strictly four prevailing transport phenomena involved in drying (internal and external head transfer and internal and external mass transfer), all of which describe the drying process. Nevertheless, the corresponding four classical partial differential equations demand considerable computing time for their numerical solution (CRANK, 1975).

According to Panchariya et al. (2002), assuming that the resistance to moisture flow is uniformly distributed throughout the interior of the homogeneous isotropic material, the diffusion coefficient, D, is independent of the local moisture content. Moreover, if the volume shrinkage is negligible, Fick's second law may be derived as follows:

$$\frac{\partial M}{\partial t} = D.\nabla^2 M \quad (1)$$

Crank (1975) proposed the analytical solutions of Equation (1) for various regularly shaped bodies, such as rectangular, cylindrical and spherical items, which have been successfully predicted by using Fick's second law.

The semi-theoretical models are generally derived by simplifying general series solutions of Fick's second law or by modifying simplified models and valid within temperature, relative humidity, air flow speed and moisture content range for which they were developed. These models are listed in Table 1.

Bayesian inference

In general, the study on the evaluation of drying models has been carried out from a frequentist approach, adjusting non-linear models which aim at synthesizing pieces of information into interpretable estimates of parameters. Estimation is based on iterative processes, such as Gauss-Newton, DUD and Marquardt algorithm, due to the non-linearity of 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, the interactive methods frequently produce negative estimates for the parameters. This may cause the formation of atypical curves. Further, in the case of comparison of curves from different treatments, the distribution of non-linear model parameter estimators do not usually follow Gaussian distribution. Therefore, the process to formulate statistical tests becomes complex when the presuppositions related to the asymptotic theory are not attended (OLIVEIRA et al., 2012a). 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., 2009, 2011, 2012a and b, 2013). Obviously, inferences in relation to interest parameters should only be made after the convergence analyses of chains created.

Current research aimed at (1) observing the effect of process parameters such as drying temperature and air-flow rate; (2) estimating drying constants of some drying models by Bayesian inference.

Table 1. Mathematical models applied to drying curves.

Model	Name of model	Equation	Restrictions
Equation 2	Lewis	$y = MR = e^{-kt}$	k > 0
Equation 3	Henderson and Pabis	$y = MR = ae^{-kt}$	a > 0, k > 0
Equation 4	Logarithm	$y = MR = ae^{-kt} + c$	a > 0, k > 0, c > 0
Equation 5	Page	$y = MR = e^{-kt^n}$	k > 0, m > 0
Equation 6	Modified Page	$y = MR = ae^{-kt^n}$	a > 0, k > 0, m > 0
Equation 7	Overhults	$y = MR = e^{(-(kt)^n)}$	k > 0, m > 0
Equation 8	Modified Overhults	$y = MR = ae^{(-(kt)^n)}$	a > 0, k > 0, m > 0
Equation 9	Wang and Singh	$y = MR = 1 + at + bt^2$	a, b real
Equation 10	Thompson	$t = a \ln(MR) + b [\ln(MR)]^2$	a, b real

Source: Adapted from Faria et al. (2012).

Material and methods

Passion fruit seeds

A local industry provided passion fruit seeds. They were washed to remove juice leftover and the cleaned seeds were stored in a freezer at -4ºC, until the experiments were performed. Seed moisture contents were obtained by the Rules for Analyses of Seeds (BRASIL, 1992) at 105 ± 1ºC for 1 hour prior to the beginning of the experiments.

The laboratory dryer

Drying experiments were performed in a laboratory scale hot-air dryer. The dryer consists basically of a centrifugal fan, electric heaters and temperature indicator. Air temperature was controlled by a proportional controller and air speed by a diaphragm system. The air was heated while flowing through electric heating elements. Samples were dried in a rectangular basket with a flow cross-section of 45 x 44 cm. Air flow was perpendicular to passion fruit seeds.

Experimental procedure

The experiments were performed with only two factors (temperature and air speed), and each factor was performed at two levels (temperature at 40 and 60ºC; air speed at 0.6 and 0.9 m s^{-1}) in a 2^2 factorial design.

The 500 g sample was uniformly spread in a basket on a single layer after the desired drying conditions had stabilized. Water losses were measured by weighing the basket and its content at 3 min. intervals, until constant weight. These tests were performed in duplicate and averages were reported and used in simulation modeling.

Modeling with Bayesian inference

For the first stage it was assumed that MRs have normal distribution in the range of [0,1], i.e:

$$MR \sim Normal\,(\mu,\sigma)_{[0,1]}$$

$E(Y) = \mu =$ proposed model. The models are shown in Table 1.

All model parameters were considered *a priori* as non-informative Gamma distribution in the range of (0,1), i.e:

$$parameters \sim Gamma\left(10^{-3},10^{-3}\right)I_{(0,1)}$$

Posterior distributions of the parameters were obtained by BRugs on R program. 2,000,000 samples were obtained by Monte Carlo Markov Chain (MCMC), where 200,000 were discarded (‘burn-in samples’) to eliminate the effect of initial values. Final samples were taken with steps of 100 which contained 18,000 obtained values. Convergence chains were verified by Convergence Diagnosis and Output Analysis - CODA program (BEST et al., 1995), by Geweke (1992) and Heibelberger and Welch (1983) criteria.

DIC (Deviance Information Criterion) was used for comparison and the selection of (co)variables in models. Spigelhalter et al. (2002) suggested the use of difference criterion module between DICs values of two models, A and B analyzed. Equation 11 demonstrates this criterion.

$$D = \left|DIC_A - DIC_B\right| \tag{11}$$

So, if D < 5, there is no significant difference; if 5 ≤ D ≤ 10, there is significant difference; if D > 10, there is highly significant difference.

Results and discussion

Influence of process parameters

Table 2 provides drying times according to the experimental conditions selected. Moisture ratio versus time curves for thin layer drying of passion fruit seeds as influenced by temperature is shown in Figure 1. As expected, air temperature affected drying curves by decreasing sample drying time and increasing drying temperature. When an increase in the drying rate occurred, drying time decreased.

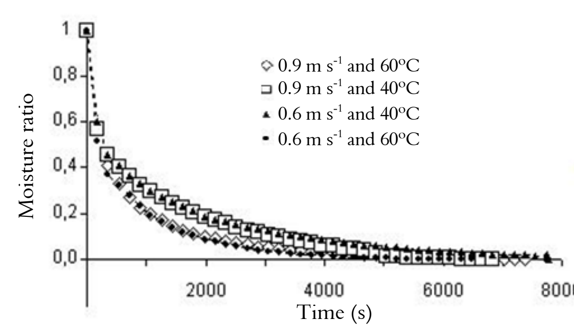

Figure 1. Variation of passion fruit seed moisture ratio with time at different temperatures and air speed.

Table 2. Drying conditions versus drying times.

Air speed (m s^{-1})	Temperature (ºC)	Drying time (s)
0.6	40	9.54
0.6	60	6.21
0.9	40	6.80
0.9	60	6.60

It may be observed that there was no constant drying rate period in passion fruit seed drying. All drying tests occurred during falling rate period and showed that diffusion was the physical mechanism underlying the process. Similar results were reported by different authors in the drying of vegetables and fruits (AZZOUZ et al., 2002; CARLESSO et al., 2005; DOYMAZ 2004a and b, 2005; ERTEKIN; YALDIZ, 2004; GOUVEIA et al., 2003; PANCHARIYA et al., 2002; SIMAL et al., 1996) Analysis of variance showed that the main effects were statistically significant only for temperature ($p < 5\%$) and that there was interaction between air-speed and temperature ($p < 5\%$).

Model fitting

In order to determine moisture content as a function of drying time, the models listed in Table 1 were fitted by Bayesian inference and DIC was obtained. DICs and estimated parameters for these models as well as average, standard deviation, credibility interval (at 95%) are presented in Tables 3 and 4.

Table 3. Bayesian estimates for model parameters for passion fruit seed drying at 0.6 m s^{-1}.

Model	Temperature 40ºC		Temperature 60ºC	
	Mean	DIC	Mean	DIC
Lewis	$k = 1.921 \times 10^{-3}$	-91.49	$k = 1.082 \times 10^{-3}$	-125,4
Henderson and Pabis	$a = 8.428 \times 10^{-1}$ $k = 1.532 \times 10^{-3}$	-99.35	$a = 7.375 \times 10^{-1}$ $k = 7.090 \times 10^{-4}$	-156.5
Logarithmic	$a = 8.412 \times 10^{-1}$ $k = 1.758 \times 10^{-3}$ $c = 2.281 \times 10^{-2}$	-103.1	$a = 7.475 \times 10^{-1}$ $k = 9.016 \times 10^{-4}$ $c = 3.380 \times 10^{-2}$	-166.2
Page	$k = 2.951 \times 10^{-2}$ $n = 5.731 \times 10^{-1}$	-188.6	$k = 3.11 \times 10^{-2}$ $n = 5.733 \times 10^{-1}$	-289.4
Modified Page	$a = 9.933 \times 10^{-1}$ $k = 3.102 \times 10^{-2}$ $n = 5.763 \times 10^{-1}$	-186.8	$a = 9.859 \times 10^{-1}$ $k = 2.777 \times 10^{-2}$ $n = 5.443 \times 10^{-1}$	-288.2
Overhults	$k = 2.421 \times 10^{-3}$ $n = 5.725 \times 10^{-1}$	-188.3	$k = 1.41 \times 10^{-3}$ $n = 5.370 \times 10^{-1}$	-289.3
Modified Overhults	$a = 9.907 \times 10^{-1}$ $k = 2.374 \times 10^{-3}$ $n = 5.784 \times 10^{-1}$	-185.5	$a = 9.877 \times 10^{-1}$ $k = 1.373 \times 10^{-3}$ $n = 5.439 \times 1^{-1}$	-288.4
Wang and Singh	$a = -5.98 \times 10^{-4}$ $b = 8.00 \times 10^{-8}$	-13.52	$a = -4.038 \times 10^{-4}$ $b = 3.716 \times 10^{-8}$	-18.92
Thompson	$a = -603.3$ $b = 48.4$	726.6	$a = -5.023 \times 10^{2}$ $b = 2.339 \times 10^{2}$	1414

By the criterion shown in Equation 11 to all conditions of air flow rate and temperature, Page, Modified Page, Overhults and Modified Overhults models were not significantly different among themselves. Thus, the above models were the best ones as they presented the smallest DICs. Page model was preferred to Modified Page, Overhults was preferred to Modified Overhults, with only two parameter to fit. Doymaz (2005, 2004b) studied the kinetics of carrots and okra respectively by frequentist approach, and concluded that Page model might be assumed to represent drying behavior.

Table 4. Bayesian estimates for model parameters for passion fruit seed drying at 0.9 m s^{-1}.

Model	Temperature 40ºC		Temperature 60ºC	
	Mean	DIC	Mean	DIC
Lewis	$k = 1.132$	-78.02	$k = 1.776 \times 10^{-3}$	-124.3
Henderson and Pabis	$a= 7.564 \times 10^{-1}$ $k = 7.886 \times 10^{-4}$	-99.48	$a =8.66 \times 10^{-1}$ $k =1.47 \times 10^{-3}$	-132.5
Logarithmic	$a = 8.474 \times 10^{-1}$ $k = 1.511 \times 10^{-3}$ $c = 2.742 \times 10^{-2}$	-127.3	$a = 8.721 \times 10^{-1}$ $k = 1.751 \times 10^{-3}$ $c = 2.942 \times 10^{-2}$	-146.1
Page	$k = 2.671 \times 10^{-2}$ $n = 5.559 \times 10^{-1}$	-161.8	$k = 2.716 \times 10^{-2}$ $n = 5.57 \times 10^{-1}$	-288.7
modified Page	$a = 9.782 \times 10^{-1}$ $k = 2.434 \times 10^{-2}$ $n = 5.665 \times 10^{-1}$	-161.1	$a =8.287 \times 10^{-1}$ $k =1.99\ 0x\ 10^{-2}$ $n =5.00 \times 10^{-2}$	-288.5
Overhults	$k = 1.482 \times 10^{-3}$ $n= 5.543 \times 10^{-1}$	-161.5	$k = 2.147 \times 10^{-3}$ $n = 5.875 \times 10^{-1}$	-288
Modified Overhults	$a = 9.824 \times 10^{-1}$ $k = 1.431 \times 10^{-3}$ $n = 5.60 \times 10^{-1}$	-160.2	$a = 1.002 \times 10^{-0}$ $k = 2.151 \times 10^{-3}$ $n = 5.874 \times 10^{-1}$	-285.3
Wang and Singh	$a = -4.952 \times 10^{-4}$ $b = 5.772 \times 10^{-8}$	-25.03	$a = -5.008 \times 10^{-4}$ $b = 5.562 \times 10^{-8}$	-13.31
Thompson	$a = -5.478 \times 10^{2}$ $b = 1.008 \times 10^{2}$	1057	$a = -4.721 \times 10^{2}$ $b = 1.53 \times 10^{2}$	763.4

Page model parameters did not change significantly to show temperature dependence, and values may be considered constant and equal to 5.5 and 5.7, respectively to 0.6 and 0.9 m s^{-1}. Senadeera et al. (2003) and Simal et al. (2005) observed the same behavior with regard to the drying of kiwi and beans, potato and peas, respectively. Azzouz et al. (2002) reported that n parameter was a function of air flow rate and initial conditions of moisture. Karathanos and Belessiotis (1999) proposed n parameter rates between 1.02 and 1.79, depending on product type. According to these authors, n rates increased with the existence of an outer skin of the dried product being higher as skin thickness increased.

However, k parameter of Page model increased with temperature rise. Simal et al. (2005) and Azzouz et al. (2002) obtained similar results.

Calculation of moisture diffusivity and activation energy

A constant rate-period was not observed in any experiments in current research and diffusion was generally accepted to be the main mechanism during the transport of moisture to the surface to be evaporated. According to Crank (1975), the solution of Fick's second law (Equation 12) out of spheres could fit the experimental drying data:

$$MR = \frac{M - M_e}{M_o - M_e} = \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} \exp\left(-\frac{n^2 \pi^2 D_{eff} t}{r^2} \right) \quad (12)$$

According to several authors (DOYMAZ; PALA, 2002; NUH; BRINKWORTH, 1997; PALA et al., 1996; PARK et al., 2001), Equation (12) assumes that the effective diffusivity (D_{eff}) is constant and sample shrinkage is negligible. For long drying time (setting n = 1), it was demonstrated that Equation (12) could be further simplified to a straight-line equation as:

$$\ln(MR) = \ln\left(\frac{6}{\pi^2}\right) - \left(\frac{\pi^2 D_{eff}}{r^2} t\right) \quad (13)$$

The effective diffusivity was calculated by Equation (13) using slopes derived from linear regression of ln(MR) against time data shown in Figure 2. An effective diffusivity is generally employed due to limited information on the movement mechanism during drying and to the complexity of the process.

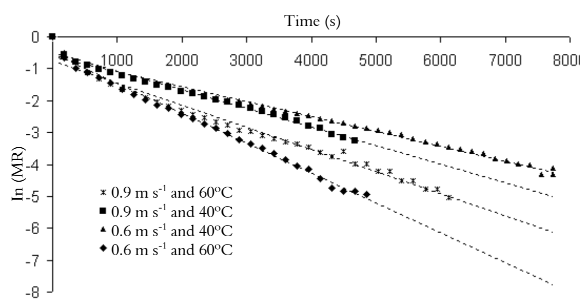

Figure 2. Experimental and predicted logarithmic moisture ratio at different drying time conditions.

Effective diffusivities (D_{eff}) during passion fruit seed drying were: 1.65 x 10^{-10} m^2 s^{-1} at 0.9 m s^{-1} and 60ºC; 1.19 x 10^{-10} m^2 s^{-1} at 0.9 ms^{-1} and 40ºC; 1.14 x 10^{-10} m^2 s^{-1} at 0.6 m s^{-1} and 40ºC, and 1.55 x 10^{-10} m^2 s^{-1} at 0.6 m s^{-1} and 60ºC. According to Mandamba et al. (1996), in the case of food materials, rates lie within the general range of 10^{-11} m^2 s^{-1} to 10^{-9} m^2 s^{-1}. By increasing the temperature, effective diffusivity(D_{eff}) increased at constant air speed, showing that there was a reduction of drying internal resistance with temperature rise. Similar behavior was reported by Panchariya et al. (2002) to black tea; Doymaz (2004a) to carrots; Doymaz (2004b) to white mulberry; Doymaz (2005) to okra. The effective diffusivity also increased when air speed rose. In fact, increase was more pronounced when the speed of drying air equaled 0.9 m s^{-1}. According to Park et al. (2001), this behavior may be explained by a decrease in external resistance when the convection coefficient and drying air temperature speed increased. Convection increase caused an increase in the material temperature and temperature rise reduced the internal resistance.

Rizvi (1986) reported that effective diffusivities depended on the drying air temperature and on material variety and composition. Sorption heat, which is a measure of moisture mobility within the food, is another factor that affected effective diffusivity. Effect of temperature on effective diffusivity was described by using Arrhenius relationship to obtain better agreement to predict curve with experimental data (CARLESSO et al., 2005; CRISP; WOODS, 1994; OZDEMIR; DEVRES, 1999).

To obtain temperature influence on effective diffusivity, rates of ln(D_{eff}) were plotted versus 1/T, as shown in Figure 3. The plot was found to be essentially a straight line in the range of temperatures investigated, indicating Arrhenius dependence (Equation 14), where E_a is the activation energy (kJ mol^{-1}), D_O is the pre-exponential factor (m^2 s^{-1}), T is the absolute temperature (K), and R is the gas constant (kJ mol K^{-1}).

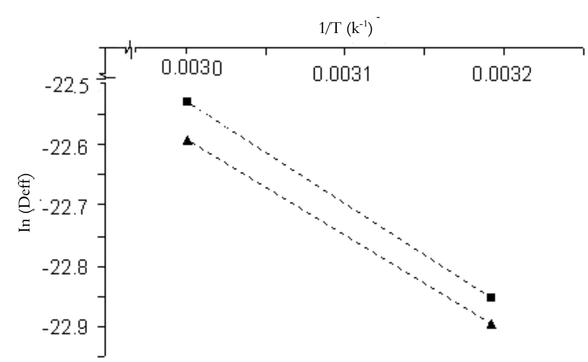

Figure 3. Effect of temperature on moisture diffusivity in passion fruit seeds.

$$D_{eff} = D_0.\exp\left(-\frac{E_a}{RT}\right) \quad (14)$$

Drying activation energy may be defined as the minimum energy required for starting diffusion through seed porous. Figure 3 shows the activation energy calculated from the straight line slope. Activation energy values were 12.8 and 19.8 kJ mol^{-1} for speed 0.6 and 0.9 m s^{-1}, respectively. Table 4 shows comparisons between literature rates and several fruits and vegetables. Pre-exponential factors were found in the same way as activation energy and were as follows: 1.90 x 10^{-8} m^2 s^{-1} and 2.75 x 10^{-8} m^2 s^{-1} to s of 0.6 m s^{-1} and 0.9 m s^{-1}, respectively. Panchariya et al. (2002) reported pre-exponential rate for the drying of black tea equals 1.68 x 10^{-7} m^2 s^{-1}.

Table 4. Activation energies of passion fruit seeds and other products.

Product	Ea (kJ mol^{-1})	References
Carrot	28.36	Doymaz (2004a)
Green peas	28,40	Simal et al. (1996)
Green beans	39.47	Senadeera et al. (2003)

Conclusion

The effects of experimental drying conditions on passion fruit seeds were studied. In the case of passion fruit seeds, drying process took place only in the falling rate period. Page, Modified Page, Overhults and Modified Overhults models may be used to describe drying behavior, although the Page model fitted better than the others. Calculated effective diffusivity rates increased with temperature rise and ranged between 1.14 x 10^{-10} and 1.80 x 10^{-10} m^2 s^{-1}. Diffusivity coefficient dependence with temperature was described by the Arrhenius-type relationship. The activation energy decreased with air flow rate.

Nomenclature

a, b, c, k, m	Constant models
D_o	Pre-exponential factor of Arrhenius equation (m^2 s^{-1})
D_{eff}	Effective diffusivity (m^2 s^{-1})
E_a	Activation energy (kJ mol^{-1})
M	Moisture constant (g water g^{-1} dry matter)
M_e	Equilibrium moisture constant (g water g^{-1} dry matter)
M_o	Initial moisture constant (g water g^{-1} dry matter)
MR	Moisture ratio
n	Positive integer
R	Gas constant (kJ mol^{-1} K^{-1})
r	Radius of passion fruit seed samples (m)
T	Temperature (K)
t	Drying time (s)

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