EVALUATION OF SOME THIN-LAYER DRYING MODELS

OCENA NEKIH MODELA KINETIKE SUŠ**ENJA**

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ABSTRACT

In this paper, a new approach for selection of a thin-layer drying model based on serial statistical criteria was proposed. The experimental data set of thin-layer drying kinetics of apple slices at five different drying air temperatures (40, 60, 80, 100 and 120 ^o C) and three drying air velocities $(1, 2 \text{ and } 3 \text{ ms}^{-1})$ were obtained on the experimental setup, designed to imitate an industrial convective *dryer. The drying data were fitted with twenty-three published thin-layer drying models (Lewis, Page, Modified Page, Henderson and Pabis, Modified Henderson and Pabis, Logarithmic(Asymptotic), Two term, Two term exponential, Midilli, Diffusion approach, Wang and Singh, Weibull, Aghbashlo, Verma., Grigoriaş, Parabolic, Hll, Demir, Logarithmic,Logistic, Vega-Lemus, Jena-Das and Alibas). In order to find which model gives the best results, some numerical experiments were made. For each model and data set, the performance index was calculated and models were ranked afterwards. After that, several statistical rejection criteria were checked. The performed analysis shows that the Alibas model could satisfactory describe the drying curve of an apple. Key words: drying, apple, thin-layer drying models, statistical criteria.*

REZIME

U ovom radu predložen je novi pristup za izbor modela kinetike sušenja na osnovu serije statističkih kriterijuma. Eksperimentalni podaci kinetike sušenja listića jabuke za pet različitih temperatura vazduha sušenja (40, 60, 80, 100 i 120°C) i tri brzine vazduha su*šenje (1, 2 i 3 ms-1) dobiveni su na eksperimentalnoj aparaturi koja je dizajnirana da imitira industrijsku konvektivnu sušaru. Aproksimacija eksperimentalnih podataka kinetike sušenja izvršena je pomoću nekoliko publikovanih modela (Lewis, Page, Modified Page, Henderson and Pabis, Modified Henderson and Pabis, Logarithmic(Asymptotic), Two term, Two term exponential, Midilli, Diffusion approach, Wang and Singh, Weibull, Aghbashlo, Verma., Grigoriaş, Parabolic, Hll, Demir, Logarithmic,Logistic, Vega-Lemus, Jena-Das i Alibas). U cilju pronalaženja modela koji najbolje aproksimira eksperimentalne podatke realizovani su numerički eksperimenti. Za svaki model i set podataka, proračunat je performansni indeks a zatim su modeli rangirani. Nakon toga proveravano je nekoliko statističkih kriterijuma. Izvršena analiza pokazuje da Alibas model sa zadovoljavajućom tačnošću opisuje krive kinetike sušenja listića jabuke.*

Ključne reči: sušenje, jabuka, modeli kinetike sušenja, statistički kriterijumi.

INTRODUCTION

The drying of food materials is a complex process of simultaneous heat and mass transfer within dried material and from its surface to the surroundings caused by a number of transport mechanisms. There are several different methods of describing the complex simultaneous heat and moisture transport processes within drying material. In the approach initially proposed by Philip and De Vries and Luikov the moisture and temperature fields in the drying material are described by a system of two coupled linear partial differential equations (Kanevce et al., 2007). On the other hand, thin-layer drying models are important tools in mathematical modeling of drying processes. They are often used to estimate drying time and generalize drying curves, and have wide application due to their ease of use and requirement of less data unlike in complex models.

Several researchers have investigated the drying kinetics of various food materials in order to determine the best mathematical model for describing thin layer drying, such as drying of apple (*Sacilik and Elcin, 2005; Hamdami et al., 2006; Meisamiasl et al., 2009; Seiiedlou et al., 2010; Grigoriaş et al., 2011; Toujani et al., 2011*), potato (*Doymaz, 2011; Thao and Noomhorm, 2011; Olawale and Omole, 2012; Shekofteh et al., 2012*) and other agriculture materials (*Yadollahinia et al., 2008; Aghbashlo et al., 2009, Taheri-Garavand et al., 2011; Doymaz* *et al., 2011a*). The thin-layer drying models which describe the drying rate of food materials are categorized into three groups: theoretical, semi-theoretical and empirical (*Erebay and Icier, 2009*).

The theoretical approach is concerned with diffusion or simultaneous heat and mass transfer equations. The semitheoretical approach is concerned with approximated theoretical equations. Empirical equations are easily applied to drying simulation as they depend only on experimental data. Theoretical approaches take into account the internal resistance to moisture transfer, while semi-theoretical and empirical approaches consider only the external resistance to moisture transfer between the product and air (*Akpinar and Bicer, 2005*).

In scientific literature generally for evaluation of thin-layer models, that use approximation of experimental drying data, the values of the coefficient of determination or R^2 or modelling efficiency EF= R^2 , correlation coefficient r, reduced chi-square χ^2 , root mean square error RMSE, and mean bias error MBE are the most common criteria to select the best model. One interesting approach for selection of the thin-layer model based on five statistical criteria was proposed from Soares et al., (*Soares et al., 2007*). In this paper a new approach for selection of a thin-layer drying model based on serial statistical criteria was proposed.

Nomenclature

MATERIALS AND METHODS

Fresh apples (Golden Delicious) were used in this study. To prepare samples, apples were washed, peeled and sliced using electric slicing machine to give a uniform sample thickness of 3 mm before being reduced to a cylinder form with diameter of 40±0.1 mm. Several measurements were made using a calliper and only samples with a tolerance of \pm 5 % were used.

The experimental data set of thin-layer drying kinetics of apple slices at drying air temperatures $40\div 120$ °C, with increment of 20° C and three drying air velocities (1, 2 and 3 ms⁻¹) were obtained on the experimental setup, designed to imitate an industrial convective dryer (Fig. 1).

Fig. 1. Experimental apparatus 1-material, 2-shelf, 3-electrical heaters,4-transformers, 5-thermocouples, 6-centrifugal fan,7-anemometer, 8-panel meter, 9-data acquisition system, 10-stove, 11-balance, 12-hygrometer

The slices have been in contact with the drying air from top and bottom surfaces. Two shelves, (Fig. 2), each holding three moist potato slices have been introduced into the rectangular experimental channel with dimensions 25x200 mm. A microthermocouple was inserted in the mid-plane of each of the three slices on the first shelf. An arithmetical mean of the readings from the three thermocouples was used as a transient temperature reading. The slices on the second shelf were weighed every ten minutes in order to obtain the volume-averaged moisture content change during drying. The temperature of the drying air, t_a , temperature of slices, t_s , has been recorded as well. The initial moisture content, M_0 , and the initial slices thickness, $2L_0$, were measured for each of the experiments (*Kanevce et al., 2007*). The initial moisture content of fresh slices and the final moisture content of dried samples were determined by hot air oven method at 105ºC for 24 h. The initial moisture content of potatoes slice was obtained as 4.81÷6.86 kg/kg d.b. Moisture content was measured by the gravimetric method using an electronic balance with precision of 0.01 g.

Fig. 2. Scheme of the drying experiment

Mathematical modeling of the drying curves

In these paper twenty-three thin-layer mathematical models, were used to approximate experimental data of the drying kinetic of apple slices (Table 1).

Table 1. Thin-layer mathematical drying models

Model	Name of model	Equation	References	
M ₀₁	Lewis (Newton)	$MR = \exp(-k_1 \tau)$	Erbay et al., 2009	
M ₀₂	Page	$MR = \exp(-k_1 \tau^m)$	Erbay et al., 2009	
M ₀₃	Modified Page	$MR = \exp[-(k_1 \tau)^m]$	Erbay et al., 2009	
M ₀₄	Henderson - Pabis	$MR = Aexp(-k_1\tau)$	Erbay et al., 2009	
M ₀₅	Modified Henderson - Pabis	$MR = Aexp(-k_1\tau)$ + $Bexp(-C\tau) + Dexp(-G\tau)$	Erbay et al., 2009	
M06	Logarithmic (Asymptotic)	$MR = Aexp(-k_1\tau) + B$	Erbay et al., 2009	
M07	Two term	$MR = Aexp(-k_1\tau)$ + Bexp($-k_2\tau$)	Erbay et al., 2009	
M08	Two term exponential	$MR = Aexp(-k_1\tau)$ + $(1-A)exp(-k1A\tau)$	Erbay et al., 2009	
M09	Midilli	$MR = Aexp(-k_1\tau^m) + B\tau$	Midilli et al., 2002	
M10	Diffusion approach	$MR = Aexp(-k_1\tau)$ + $(1-A)exp(-k1B\tau)$	Erbay et al., 2009	
M11	Wang and Singh	$MR = 1 + A\tau + B\tau^2$	Erbay et al., 2009	
M12	Weibull	$MR = \exp[-(\tau/A)^{B}]$	Doymaz, 2012	
M13	Aghbashlo	$MR = \exp[-k_1 \tau/(1+k_2 \tau)]$	Aghbashlo et al., 2009	
M14	Verma	$MR = Aexp(-k_1\tau)$ + $(1-A)exp(-B\tau)$	Verma et al., 1985	
M15	Grigoras	$MR = A(1-\exp(-B\tau))$	Grigoraș et al., 2011	
M16	Parabolic	$MR = A+B\tau+C\tau^2$	Doymaz, 2011	
M17	Hll	$MR = Aexp(-k_1\tau^m)$ + Bexp($-C\tau^m$)	Hll et al., 2011	
M18	Demir	$MR = Aexp(-k_1\tau)^m + B$	Erbay et al., 2009	
M19	Logarithmic	$MR = A+Bexp(-k_1\tau)$	Cihan et al., 2007	
M20	Logistic	$MR = A/[1+Bexp(k_1\tau)]$	Cihan et al., 2007	
M21	Vega-Lemus	$MR = (A + k_1 \tau)^2$	Cruz et al., 2012	
M22	Jena and Das	$MR = Aexp(-k_1 \tau + B \tau^{0.5}) + C$	Jena et al., 2017	
M23	Alibas	$MR = Aexp[(-k_1\tau^m)+B\tau] + C$	Alibas, 2012	

In these models, the moisture ratio (MR) is defined by the following equation

$$
MR = (M - M_{eq}) / (M_0 - M_{eq})
$$
 (1)

The values of M_{eq} are relatively little compared to those of M or M_0 , so the error involved in the simplification is negligible. Thus moisture ratio was calculated as:

$$
MR = M / M_0
$$
 (2)

In order to estimate and select the best thin-layer drying model several statistical criteria were used The value of performance index, φ which is calculated on the basis of calculated values for coefficient of determination \mathbb{R}^2 , the root mean squared error RMSE and the mean relative deviation MRD is the first statistical criteria for selection of thin-layer drying model (*Ruiz-Lopez and Lara, 2009*):

$$
R^{2} = \frac{SS_{M} - SS_{E}}{SS_{M}}
$$
, $RMSE = \sqrt{MS_{E}}$, $MRD = \frac{1}{n} \sum_{i=1}^{n} \left| \frac{E_{i}}{MR_{e}} \right|$ (3)

$$
MS_{_E} = \frac{SS_{_E}}{n-k}\,,\;\; SS_{_M} = \sum_{i=1}^n \bigl(MR_{_E} - \bar{MR}_{_e}\bigr)^2\,\,,\; SS_{_E} = \sum_{i=1}^n E_i^{\,2}\,\,,
$$

$$
E_i = MR_e - MR_e, \overline{MR}_e = \frac{1}{n} \sum_{i=1}^{n} MR_e
$$
 (4)

$$
\phi = \frac{R^2}{RMSE \cdot MRD} \tag{5}
$$

Higher values of performance index ϕ indicate that thin-layer model better approximates the experimental data.

The D'Agostino-Pearson test of normality is the most effective procedure for assessing a goodness of fit for a normal distribution (Sheskin, 2000). This test is based on the individual statistics for testing of the population of skewness z_1 and kurtosis $z₂$, respectively. This test is second statistical criteria as adequate of thin-layer model and it is based on the following quantities (Sheskin, 2000; Ruiz-Lopez and Lara, 2009):

$$
SE_{m} = \sum_{i=1}^{n} \varepsilon_{i}^{m} \text{ for } m = 2, 3, 4, \varepsilon_{i} = E_{i} - \bar{E}, s = \sqrt{\frac{SE_{2}}{n-1}}
$$
(6)

$$
m_3 = \frac{nSE_3}{(n-1)(n-2)}, \ m_4 = \frac{n(n+1)SE_4}{(n-1)(n-2)(n-3)} - \frac{3SE_2^2}{(n-2)(n-3)}
$$
(7)

$$
A_1 = \frac{m_3(n-2)}{s^3 \sqrt{n(n-1)}} \sqrt{\frac{(n+1)(n+3)}{6(n-2)}}, \ B_1 = \frac{3(n^2+27n-70)(n+1)(n+3)}{(n-2)(n+5)(n+7)(n+9)} \tag{8}
$$

$$
C_1 = \sqrt{2(B_1 - 1)} - 1, D_1 = \sqrt{C_1}, E_1 = \frac{1}{\sqrt{\ln(D_1)}}, E_1 = \frac{A_1 \sqrt{C_1 - 1}}{\sqrt{2}} \tag{9}
$$

$$
G_2 = \frac{24n(n-2)(n-3)}{(n+1)^2(n+3)(n+5)}, H_2 = \frac{(n-2)(n-3)|m_4|}{s^4(n-1)(n+1)\sqrt{G_2}}
$$
(10)

$$
J_2 = \frac{6(n^2 - 5n + 2)}{(n + 7)(n + 9)} \sqrt{\frac{6(n + 3)(n + 5)}{n(n - 2)(n - 3)}},
$$

\n
$$
K_2 = 6 + \frac{8}{J_2} (\frac{2}{J_2} + \sqrt{1 + \frac{4}{J_2^2}}), \quad L_2 = \frac{1 - \frac{2}{K_2}}{1 + H_2 \sqrt{\frac{2}{K_2 - 4}}}
$$
(11)

$$
z_1 = E_1 \ln(F_1 + \sqrt{F_1^2 + 1}) \cdot z_2 = \left(\frac{2}{9K_2}\right)^{-\frac{1}{2}} \left(1 - \frac{2}{9K_2} - \sqrt[3]{L_2}\right) \tag{12}
$$

The test statistic for the D'Agostino-Pearson test of normality is computed with equation

$$
\chi^2 = z_1^2 + z_2^2 \tag{13}
$$

The χ^2 statistics has a chi-squared distribution with 2 degrees of freedom (df). The tabled critical .05 chi-square value for $df =$ 2 is $\gamma_{.05}^2$ = 5.99. Therefore, if the computed value of chi-square is equal to, or greater than, either of the aforementioned values, the null hypothesis can be rejected at the appropriate level of significance (*Sheskin, 2000*), i.e. the thin-layer model should be rejected.

Because the χ^2 statistics is not recommended individually as an adequate measure of the effectiveness of a thin-layer model to describe the experimental data, additional criterion has to be introduced. The single-sample run test is one of them and it is third statistical criteria adequate of thin-layer model. The singlesample run test is one of a number of statistical procedures that have been developed for evaluating whether or not the distribution of series is random. The test evaluates the number of runs in a series in which, on each trial, the outcome must be one of $k = 2$ alternatives. In this test the number of positive and negative residuals (n_1 and n_2 , respectively) and the number of times the sequence of residuals changes sign (g) are used to calculate the following test statistic (Sheskin, 2000):

$$
z_r = \frac{|g - g_1| - 0.5}{\sigma_r}
$$
 (14)

$$
g_1 = \frac{2n_1n_2}{n_1 + n_2} + 1 \cdot \sigma_r = \sqrt{\frac{2n_1n_2(2n_1n_2 - n_1 - n_2)}{(n_1 + n_2)^2(n_1 + n_2 - 1)}}
$$
(15)

The standardized variable z_r has a normal distribution with zero mean and unit variance. A two-sided test must be performed to account for too few or too many runs. Thus, if the computed value of z_r is greater than the tabled critical two-tailed value z_{05} = 1.96 the null hypothesis should be rejected (Sheskin, 2000), i.e. the thin-layer model should be rejected.

A fourth statistical criterion for selection of thin-layer drying model is the significance and precision of the model constant. That can be done with constructing of individual confidence intervals (CI). If the estimated value of parameters is out of the 95% confidence interval, the model contains irrelevant parameters for approximation of experimental data i.e. the thin-layer model should be rejected.

RESULTS AND DISCUSSIONS

The experimental data of drying kinetic of apple slices were fitted to twenty three thin-layer mathematical models by means of nonlinear regression analysis using **nlinfit** function of the Statistic Toolbox of Matlab 7.1 (The MathWorks Inc., Natick, MA, USA). On the basis of experimental data, and each model from Table 1, the average values of: coefficient of determination R, root mean squared error RMSE, the mean relative deviation MRD and performance index ϕ were calculated. After that, models were ranked on the basis of average values of performance index ϕ_{av} (Table 2). From Table 2, it is evident that models M23, M16, M11, M13, M09 and M20, have higher value of average performance index ϕ_{av} compared with the other models. The model M23 i.e. model of Alibas has the highest value of average performance index ϕ_{av} = 11017.670, while the models M15 i.e. the model of Grigoraş, have the smallest value of average performance index $\phi_{av} = 1.803$. In Table 3, the computed average values for χ^2 and z_r are given.

It is obvious that the models M11 and M20, have average value of z_r higher than the tabled critical two-tailed value $z_{.05}$ = 1.96, although have higher performance index. In accordance with the third statistical criteria, these models were rejected in further consideration. For models M23, M16, M13 and M09 computed average value of chi-square χ^2_{av} and z_{rav} are smaller than critical value. In accordance with statistical criteria, those models are able to correlate the experimental values of drying kinetic of apple slices with $0.4 \div 0.8$ % average root mean squared error. But, from the first statistical criteria, the model M23 i.e. Alibas model have highest values of performance index φ than models M16, M13 and M09, so that model the best approximate the experimental data.

The estimated values of parameters and 95% confidence intervals of estimated parameters for the model M23, for all air drying temperatures and air drying velocities are given in Table 4. The 95% confidence intervals of the estimated parameters were determined by using the nlparci function of the same Matlab Toolbox. As shown in Fig. 3, a good match was found between experimental and calculated values with the model of Alibas.

Fig. 3. Experimental and predicted moisture ratio for different air temperatures and velocities

Analyzing the residues of the model M23, the plots of the residues against the experimental values did not indicate abnormal distribution (not presented here).

$t[^oC]$ v $\rm [ms^{-1}]$	A 95% CI	\mathbf{k}_1 95% CI	m 95% CI	В 95% CI	$\mathbf C$ 95% CI
	-0.001	-3.851	0.158	-0.005	1.011
40	$(-1E-3,$	$(-6.142,$	(0.098,	$(-0.006,$	(1.001,
1	1E3)	$-1.561)$	0.217)	-0.005)	1.020)
40	$-1.2E-05$	-7.192	0.106	-0.007	1.009
2	$(-8)E-05$	(-12.644)	(0.047,	$(-0.008,$	(1.00,
	$, 6E-05)$	$, -1.739)$	0.166)	-0.006	1.017)
40	-0.001	-4.326	0.141	-0.007	1.015
3	$(-0.002,$	$(-8.386,$	(0.050,	$(-0.008,$	(1.001,
	0.001)	-0.266	0.232)	$-0.005)$	1.030)
60	-0.001	-2.973	0.214	-0.014	1.009
1	$(-0.004,)$	$(-4.604,$	(0.143,	$(-0.016,$	(0.999,
	0.001)	$-1.342)$	0.286)	-0.012)	1.019)
60 2	-0.001	-3.206	0.199	-0.013	1.008
	$(-0.004,)$	$(-5.064,$	(0.127,	$(-0.015,$	(0.998,
	0.001)	-1.347	0.272)	$-0.011)$	1.018)
60	-0.002 $(-0.008,$	-3.203	0.188	-0.013	1.008
3	0.005)	$(-6.375,$ $-0.031)$	(0.065, 0.3108)	$(-0.016,$ -0.009	(0.992, 1.024)
	-0.001	-3.338	0.211	-0.019	1.008
80	$(-0.004,)$	$(-5.877,$	(0.108,	$(-0.023,$	(0.996,
1	0.0019	-0.799	0.314)	$-0.015)$	1.020)
	-0.002	-3.223	0.217	-0.023	1.007
80	$(-0.005,$	$(-5.543,$	(0.115,	$(-0.028,$	(0.995,
2	0.003)	$-0.902)$	0.3188)	-0.018	1.018)
	-0.001	-3.330	0.227	-0.028	1.007
80	$(-0.007,$	$(-7.513,$	(0.038,	$(-0.039,$	(0.990,
3	0.004)	0.854)	0.415)	-0.016	1.024)
	-0.001	-3.740	0.206	-0.022	1.003
100 1	$(-0.004,)$	$(-8.394,$	(0.033,	$(-0.031,$	(0.989,
	0.002)	0.914)	0.378)	$-0.012)$	1.016)
100	-0.002	-3.030	0.245	-0.034	1.007
2	$(-0.016,$	$(-9.023,$	$(-0.080,$	$(-0.060,$	(0.979,
	0.012)	2.964)	0.569)	-0.009	1.034)
100	-0.002	-3.039	0.236	-0.033	1.003
3	$(-0.009,$	$(-5.353,$	(0.115,	$(-0.042,$	(0.991,
	0.004)	$-0.726)$	0.358)	$-0.024)$	1.014)
120	-0.002	-3.247	0.238	-0.038	1.006
1	$(-0.001,$	$(-7.221,$	(0,0398)	$(-0.054,$	(0.988,
	0.006) -0.003	0.727) -2.472	0.435) 0.316	$-0.021)$ -0.055	1,023) 1.006
120	$(-0.031,$	$(-9.64,$	$(-0.285,$	$(-0.136,$	(0.966,
2	0.025)	4.700	0.917)	0.255)	1.046)
	-0.003	-2.909	0.2723	-0.057	1.005
120	$(-0.034,$	(-10.782)	$(-0.245,$	$(-0.123,$	(0.966,
3	0.027)	4.963)	0.790)	0.008	1.044)

Table 4. Non-linear regression parameters and 95% CI

CONCLUSION

A single statistical criterion cannot be used to select the thinlayer drying model. The model must always be chosen based on multiple statistical criteria. For this reason, a series of statistical criteria for choosing of best thin-layer model were proposed. Twentythree existing thin-layer drying models were applied to approximate experimental drying data of the drying kinetic of apple slices. From third rejection criterion, it is obvious that little models have average values of z_r lower than the tabled critical value (only four from

twenty-three). Those models are an accurate tool for approximation of the experimental drying data. But, based on the results on the first statistic criterion, highest value of average performance index φ, have Alibas model. This exhibited that this model has the best ability to correlate the experimental drying data. This model is able to explain 99.9% of the variation of MR when the experimental conditions and moisture content of drying material are known. From all models, Grigoraş and Two term exponential model exhibited the worst statistical results.

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