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Drying characteristics of walnut (*Juglans regia* L.) during convection drying**

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A b s t r a c t. Drying characteristics of walnut were determined experimentally as a function of temperature, air velocity and variety (Serr, Pedro, Z67, K82). Four different drying models were fitted to the experimental data to select a suitable mathematical form of the drying curve. Experiments were performed at air temperatures of 32 and 43°C and two air velocities of 1 and 3 m s^{-1} at each temperature. Of the drying models tested, the Page model was selected as the best mathematical model based on the values of R^2, χ^2 and *RMSE*. Drying time and Page model constants were significantly dependent on the variables studied. Correlations of the Page model constant, k and m, the effective moisture diffusivity of walnuts and the drying time with the variables were determined. The effective moisture diffusivity of walnuts varied from 3.54×10^{-7} to 9.92×10^{-7} m^2 s^{-1}.

K e y w o r d s: drying, walnut, modelling, Page model, effective moisture diffusivity

INTRODUCTION

The genus Juglans includes about 20 species with a natural distribution across the Northern hemisphere and extending into South America. Most of the species are commercially important as a source of edible walnut, highly prized timber, and as specimen trees. The Persian walnut (*Juglans regia* L.) is a traditional nut of Old World agriculture and grows in mesic, temperate, deciduous forests of Balkan, North Turkey, South Caspian Region, Caucasus and Central Asia (Petrieeione and Aliotta, 2006). The walnut is mainly produced in China, USA, Iran, and Turkey. Based on FAO statistics (Food and Agriculture Organization, 2006), Iran produced about 150 Mt of walnut in 2006, which was approximately 10% of the world's walnut production. The walnut fruits are a highly nutritious food, rich in oil composed of unsaturated fatty acids. The walnut kernel represents from 40 to 60% of the nut mass, depending mainly on the variety. The kernel has high levels of oil (60-75%) in which polyunsaturated fatty acids predominate. In addition to oil, the walnut kernel provide appreciable amounts of proteins (up to 15% of the kernel mass), carbohydrates (12-16%), fibre (up to 2.1%) and minerals (1.7-2%) (Mitra *et al*., 1991).

Proper harvesting and post-harvest handling are the key to achieving maximum yield of good quality nuts that determine marketability and profit (Kashaninejad *et al*., 2007). Like other nuts and beans, drying is one of the common methods of walnut processing. Drying stabilizes the product mass, prevents deterioration in kernel quality, improves bleaching efficiency and prolongs storage life (Mitra *et al*., 1991).

Generally, the recommended temperature of hot air for drying process of walnut should be in the range of 32-43°C and drying process continued until the moisture content reaches 8% (% w.b.). Hot air temperatures out of this range darken kernels and reduce their storage life (Mitra *et al*., 1991; Fallahi, 1998). Laboratory and field experiments have been used to develop mathematical models that simulate the drying process (Fallahi, 1998). Simulation models may be used to develop new drier systems that result in energy savings and optimum processing conditions. Several researchers have investigated the drying kinetics of various nuts to find the best mathematical models for describing the drying characteristics of nuts such as pistachio nuts (Kashaninejad *et al*., 2007), hazelnuts (Ceylan and Aktas, 2008) and hazelnuts during roasting (Ozdemir and Devres, 1999). Literature survey showed that limited published data is available on walnut drying. The objectives of this study were: a) to study the effect of temperature, velocity and variety on drying kinetics of walnut; b) to select a suitable thin-layer drying model; and c) to calculate drying constants.

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MATERIALS AND METHODS

Freshly harvested walnuts with green skin (*Juglans regia* L.) were obtained from the Seed and Plant Improvement Institute (SPII) of Karaj (Iran) and stored in a refrigerator at about +5°C. The samples were transported to drying Laboratory of Abureyhan University College, Pakdasht, Iran. Generally, samples of uniform size were selected for drying experiments. The outer green skin of walnuts was removed prior to drying. The initial moisture content of walnuts was determined by the oven method. Halved walnuts were dried in an oven at 105±2°C until the mass did not change between two weighing. At least five replicates for each varieties of experiment were measured. Before the experiments, linear dimensions of each variety of walnut – length (L), width (W), and thickness (S) – were measured using a micrometer to an accuracy of 0.01 mm.

Figure 1 shows a schematic view of laboratory-scale hot-air dryer that was used for this study. Essential parts of the dryer system include: an adjustable centrifugal blower, an air heating chamber (4.5 kW), a drying chamber, a system controller unit, a frequency inverter to control motor speed (Lenze, 8300, Germany) and a sample tray. Relative humidity and temperature of ambient air around the dryer were 35 to 40% and 18 to 26°C, respectively. To decrease the undesirable effects of ambient temperature and humidity of air on the experiments, drying chambers and tunnel of the dryer were isolated with rock wool and wood. The drying tray was insulated by a glassy cylinder and hot air exhaust from upper part of the glassy cylinder. The dryer had an automatic temperature controller that worked with an accuracy of ±0.1°C. The air velocity was adjusted to the desired levels of 1.0 or 3.0 m s^{-1} using a frequency inverter which was directly acting on the blower motor (1.5 kW). Air velocity was measured with accuracy of ±0.05 m s^{-1} by Lutron PROVA AVM-07 anemometer. The dried samples were weighed by an electronic digital balance with an accuracy of 0.01 g (AND, Japan). The dryer system was started 1 hour prior to placing the walnuts in the dryer in order to achieve a steady state operating condition. Walnut masses were recorded hourly. The experiments continued until the moisture content (% w.b.) of the sample reached about 8 (% w.b.).

All drying experiments in this study were repeated three times at two levels of air temperature (32 and 43°C) and two air velocity levels (1 and 3 m s^{-1}) for four walnut cultivars. The individual and combined effects of the variables (variety, air temperature and air velocity) were analysed on drying time and Page model constants of walnut drying based on the factorial experiments (three factor completely randomised design (4×2×2×3)) using MATLAB computer program.

THEORETICAL CONSIDERATIONS

Drying curves were fitted with four different moisture ratio models – the Lewis, the Henderson and Pabis, and the Page and the approximation of diffusion model. The moisture ratio of the walnut during the drying experiments was found using Eq. (1):

$$MR = \frac{M_t - M_e}{M_0 - M_e}, \tag{1}$$

where: MR is moisture ratio (dimensionless); M_t is the moisture content an any time, M_e is equilibrium moisture content, and M_o is initial moisture content (% d.b.).

One of the oldest models used to describe drying of barley was the Lewis model. The model can be presented as (Lewis, 1921):

$$MR = \exp(-kt), \tag{2}$$

where: k is drying rate (s^{-1}), t – time of drying (s).

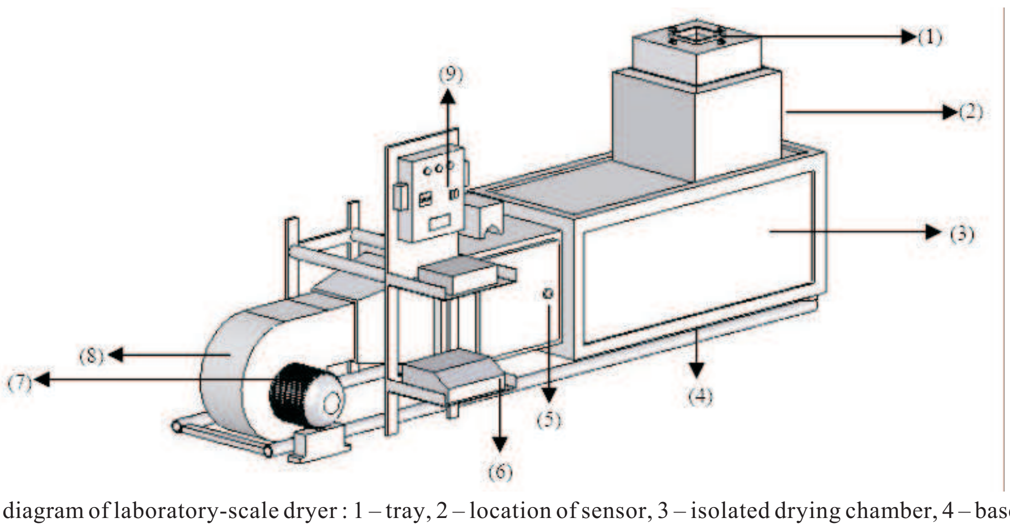

Fig. 1. Schematic diagram of laboratory-scale dryer : 1 – tray, 2 – location of sensor, 3 – isolated drying chamber, 4 – base of dryer, 5 – heater, 6 – inverter, 7 – electromotor, 8 – fan, 9 – automatic controller.

The Henderson and Pabis model is the first term of a general series solution of Fick's second law. The Henderson and Pabis model was used to model drying of maize (Henderson and Pabis, 1961). This can be written as:

$$MR = \mathrm{a}\exp(-kt), \tag{3}$$

where: a is constant (dimensionless).

Page (1949) introduced the homonymous thin-layer drying equation for shelled corn as:

$$MR = \exp(-kt^{\mathrm{m}}), \tag{4}$$

where: m is constant (dimensionless).

Approximation of diffusion model was used to model drying of grapes. The model was (Yaldiz *et al*., 2001):

$$MR = \mathrm{a}\exp(-kt) + (1-\mathrm{a})\exp(-k\mathrm{b}t), \tag{5}$$

where: b is constant (dimensionless).

Coefficient of determination (R^2), reduced chi-square (χ^2) and root mean square error (*RMSE*) were used in this study to determine the goodness of fit. The chi-square and *RMSE* were calculated using following equations:

$$\chi^2 = \frac{\sum_{i=1}^{N}(MR_{\exp,i} - MR_{pre,i})^2}{N-z}, \tag{6}$$

$$RMSE = \left(\frac{1}{N}\sum_{i=1}^{N}\left(MR_{pre,i} - MR_{\exp,i}\right)^2\right)^{\frac{1}{2}}, \tag{7}$$

where: N is number of observation, z is number of constant, MR_{exp} is experimental moisture ratio and MR_{pre} is predicted moisture ratio. The best model to describe the thin-layer drying characteristics of walnut was selected as the higher value of R^2, and lower value of χ^2 and *RMSE*.

The analytical solution of Fick's second law of unsteady state diffusion in a spherical body, proposed by Crank (1975), can describe the transport of moisture during the falling rate period by assuming that the effective moisture diffusivity is constant and radial during drying process. The effective moisture diffusivity can be calculated from the following equation:

$$MR = \frac{M_t - M_e}{M_0 - M_e} = \frac{6}{\pi^2}\sum_{n=1}^{\infty}\frac{1}{n^2}\exp\left(-n^2\pi^2\frac{D_{eff}\,t}{r^2}\right), \tag{8}$$

where: n=1,2,3,... the number of terms taken into consideration, t time of drying, D_{eff} effective moisture diffusion (m^2 s^{-1}), r_o radius of the sphere which assumed constant for the case of drying of a spherical body. For terms in the series where n>1, each term approaches zero as t increases. Neglecting higher order terms (n>1), the equation becomes:

$$MR = \frac{6}{\pi^2}\exp\left(-\pi^2\frac{D_{eff}\,t}{r^2}\right). \tag{9}$$

Eq. (9) was used by several researchers to describe the diffusion at constant radius (r) for the duration of the drying process (Babalis and Belessiotis, 2004; Doymaz, 2005; Pahlavanzadeh *et al*., 2001). Eq. (9) can be simplified to a straight-line equation as:

$$\ln(MR) = \ln\left(\frac{6}{\pi^2}\right) - \left(\pi 2\frac{De_{ff}\,t}{r^2}\right). \tag{10}$$

The diffusion coefficient is obtained by plotting experimental drying data in terms of ln(*MR*) versus time(s). From Eq. (10), a plot of ln(*MR*) versus time gives a straight line with a slope of k_1 in:

$$k_1 = \frac{\pi^2 D_{eff}}{r_g^2}. \tag{11}$$

The effective radius was determined by calculating the geometric mean diameter of the walnut, then dividing it by two (Mohsenin, 1986):

$$r_g = \frac{(LWS)^{\frac{1}{3}}}{2}, \tag{12}$$

where: r_g is the geometric radius (mm), L is the length (mm), W is the width (mm) and S is the thickness (mm).

RESULTS AND DISCUSSION

Initial moisture content (% w.b.), wood (shell) mass to total mass ratio *WR* (%) and geometric radius of fresh walnuts for each variety are reported in Table 1. The Pedro variety had the highest value of initial moisture content and wood mass to total mass ratio *WR* (%), whereas the K82 variety had the lowest value of initial moisture ratio content and wood mass to total mass ratio *WR* (%). The K82 variety had the highest value of geometric mean radius whereas the Serr variety had the lowest value of geometric mean radius.

Table 2 shows mean drying times for different drying conditions. Minimum value of drying time is 15 h for K82 variety when air velocity is 3 m s^{-1} and air temperature is 43°C. Maximum value of drying time is 34.33 h for the Pedro variety when air velocity is 1 m s^{-1} and air tempera-

Table 1. Some physical properties of fresh walnuts

Property	Pedro	Serr	Z67	K82
Initial moisture content (% w.b.)	34.03	32.51	30.25	28.45
WR (%)	59.92	52.28	50.67	47.27
Geometric mean radius (mm)	17.82	16.71	16.73	18.36

ture is 32°C. The Pedro variety had maximum value of drying time at the same value of air temperature and air velocity as the other varieties due to high value of shell fraction and initial moisture content. Whereas, reverse results were obtained for K82 variety due to low value of shell fraction and initial moisture content.

The results of analysis of variance (*ANOVA*) showed that drying air temperature, air velocity and variety had a significant effect on the drying time. Interactions of temperature and variety and temperature and velocity were not significant. However, the interaction of air velocity and variety was significant. Similar findings were reported by several researchers, based on studies in which drying air temperature is considered as the main factor affecting drying rate (Doymaz, 2004; Kashaninejad *et al*., 2007; Kashaninejad and Tabil, 2004 and Madamba *et al*., 1996).

Figure 2 shows the effect of variety on moisture ratio of walnut at air velocity of 1 m s^{-1} and air temperature of 32°C. Similar trend of moisture ratio variation was observed for different levels of air velocity and temperature. As depicted in this figure, drying of walnuts took place in one falling rate period only. It is clear from the figure that the variety had a significant effect on drying curves. Values of moisture ratio for the Pedro variety are higher than for the other varieties at each given level of time. It could be related to physical properties of Pedro variety (the highest value of W_0 and *WR* among the other varieties), but a reverse result was observed for the K82 variety, which might be due to physical properties of this variety.

The influence of air temperature on moisture ratio at air velocity of 1 m s^{-1} on Serr and Z67 varieties is shown in Fig. 3. It is clear from the figure that with increase in the drying air temperature the moisture ratio decreased because higher air temperature causes a higher reduction of moisture content. On the other hand, at high temperatures the rate of heat and mass transfer is high and water loss is excessive.

Figure 4 shows drying rate versus moisture content at air velocity of 1 m s^{-1} for Serr and Z67 varieties. It is evident that drying rate starts from zero at initial moisture content and then reaches a maximum value after a short time, then decreases with progressing time and approaches zero at the end of drying time as well as decreasing moisture content. At the same temperature, the Serr variety had a high value of drying rate because of high initial moisture content, as compared with Z67 variety. It is clear that at known variety (Serr or Z67) the high temperature caused higher drying rate.

The effect of air velocity on moisture ratio at air temperature of 32°C on Pedro and Serr varieties is depicted in Fig. 5. Similar trends were found for other temperatures and varieties. The air velocity has a significant influence on drying rate at initial time of walnuts drying due to surface moisture evaporation at initial drying time.

T a b l e 2. Effect of drying air conditions on drying time (h)

Variety	Temperature (°C)	Velocity (m s^{-1})	Drying time (h)
Pedro	43	1	31.66
	43	3	24.33
	32	1	34.33
	32	3	32.33
Serr	43	1	22.66
	43	3	22.00
	32	1	30.66
	32	3	28.66
Z67	43	1	22.66
	43	3	18.66
	32	1	27.00
	32	3	25.66
K82	43	1	16.00
	43	3	15.00
	32	1	22.00
	32	3	19.00

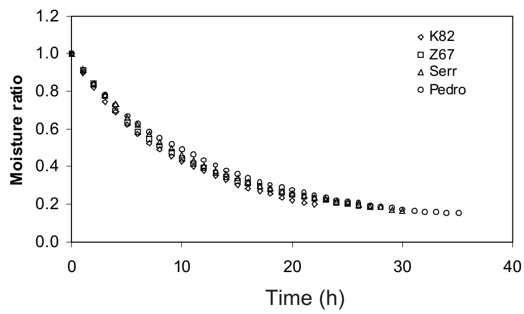

Fig. 2. Effect of variety on moisture ratio of walnuts at 32°C and V= 1 m s^{-1}.

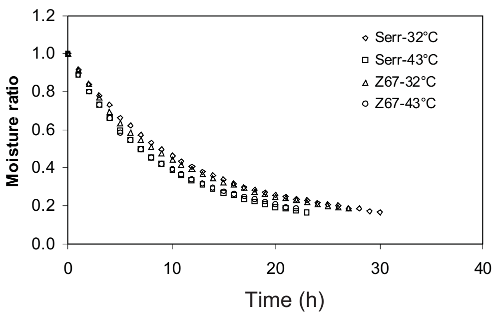

Fig. 3. Effect of temperature on moisture ratio of walnuts at V =1 m s^{-1} for Serr and Z67 variety.

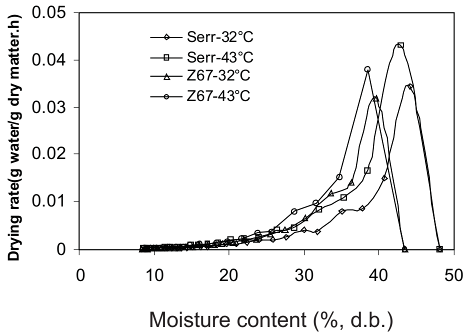

Fig. 4. Effect of temperature on drying rate of walnuts at V = 1 m s^{-1}, for Serr and Z67 variety.

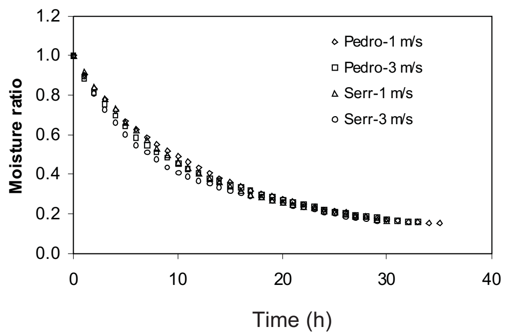

Fig. 5. Effect of air velocity on moisture ratio of walnuts at 32°C for Pedro and Serr variety.

The Lewis model, the Henderson and Pabis model, the Page model and the approximation of diffusion drying models were fitted to the drying data and sorted in descending order of R^2 and ascending order of χ^2 and *RMSE*. The results of the statistical parameter showed that the Page model was the best one for mathematical modelling of drying kinetics of walnut drying due to the highest value of R^2 and the lowest values of χ^2 and *RMSE*. Table 3 shows the fitting results of the Page model. In all of the experiments, the Page model showed the best agreement for thin-layer drying curves of walnut. Generally, R^2, χ^2 and *RMSE* values for the Page model were in the range of 0.9921-0.9998, 2.527×10^{-4}-1.029×10^{-8} and 0.002603-0.021854, respectively.

The results of analysis of variance also showed that the Page model constants were significantly influenced (P < 0.0000) by temperature, variety and air velocity (Tables 4). The results of the study presented in this paper are in agreement with the findings of Abalonel *et al*. (2006). The Page model constants were regressed against air condition using multiple regression, and the equation and related R values are reported. Regression analysis for these parameters yielded the following relationships at the statistically significant level of 1%:

$$k=\exp(-2.94+0.007531T+0.6583V+1.09203WR)$$

$$R=0.9221, \quad (13)$$

$$m=\exp(0.3252+0.003948T-0.01499V-1.2744WR)$$

$$R=0.9514. \quad (14)$$

The drying experiments were continued until the moisture content of walnuts reached to 8 (% w.b.). Figure 6 shows the ln(*MR*) versus time at four combinations of air velocities and temperatures for K82 variety. Using Eq. (11), the D_{eff} values for all of the experiments were calculated and reported in Table 5. Minimum value of moisture diffusivity is 3.54×10^{-7} m^2 s^{-1} for the Pedro variety when air velocity is 1 m s^{-1} and air temperature is 32°C. Maximum value of moisture diffusivity is 9.92×10^{-7} m^2 s^{-1} for K82 variety when air velocity is 3.0 m s^{-1} and air temperature is 43°C. The K82 variety had maximum value of moisture diffusivities at the same value of air temperature and air velocity as the other varieties, due to low value of shell fraction that providing reduced resistance to moisture movement. Therefore, a lower drying time is required to achieve given water content in K82 than in the other varieties.

Rizvi (1986) stated that effective diffusivities depend on the drying air temperature in addition to variety and composition of material. The effect of air conditions and variety physical properties on effective moisture diffusivity is generally described using Arrhenius type equation (Akpinar *et al*., 2003).

Using multiple regression analysis, the following equation was developed to model effective moisture diffusivity of walnut drying in terms of air temperature, air velocity and initial moisture content at the statistically significant level of 1%:

$$D_{eff} = 1.08\ 10^{-4} V^{0.1275} \exp\left(-\frac{2633.6}{T_{abs}}\right) \quad R=0.9193. \quad (15)$$

The drying time of walnut depends on the properties of the varieties and on the conditions of hot air drying. A multiple regression analysis was done for estimating drying time in terms of initial moisture content, air temperature and air velocity. The following equation was obtained for prediction of drying time which was highly significant. Furthermore, the coefficients of the equation were significant at the level of 1%:

$$t = \exp\left(1.3373 - 0.225T - 0.05479V + 8.905W_0\right)$$

$$R=0.9673. \quad (16)$$

T a b l e 3. Curve fitting results and constants related to the Page model

Variety	Temperature (°C)	Velocity (m s^{-1})	R^2	*RMSE*	χ^2	*k*	*m*
Pedro	43	1	0.9962	0.014031	3.964E-05	0.1561	0.7353
	43	3	0.9953	0.016278	4.753E-05	0.1740	0.7308
	32	1	0.9921	0.021019	2.527E-04	0.1398	0.7296
	32	3	0.9976	0.010974	1.674E-05	0.1465	0.7278
Serr	43	1	0.9981	0.010381	6.698E-06	0.1408	0.8188
	43	3	0.9921	0.021854	2.522E-04	0.1474	0.8068
	32	1	0.9959	0.015173	5.098E-05	0.1209	0.7994
	32	3	0.9955	0.015317	4.623E-05	0.1538	0.7379
Z67	43	1	0.9984	0.009638	4.168E-06	0.1349	0.8292
	43	3	0.9983	0.009860	3.790E-06	0.1499	0.8250
	32	1	0.9971	0.012705	1.896E-05	0.1190	0.8214
	32	3	0.9974	0.011688	1.259E-05	0.1418	0.7710
K82	43	1	0.9996	0.004718	1.263E-07	0.1287	0.9106
	43	3	0.9998	0.002603	1.029E-08	0.1477	0.8808
	32	1	0.9992	0.006402	8.115E-07	0.1197	0.8475
	32	3	0.9985	0.008922	2.541E-06	0.1452	0.8185

T a b l e 4. Analysis of variance (ANOVA) for the effect of variety, air temperature, and air velocity on *k* (Page model constant)

Source	d.f.	*MSE*	F	P>F	*MSE*	F	P>F
		k			*m*		
Temperature	1	0.00156	69.48***	0.0000	0.00158	207.54***	0.0000
Variety	3	0.00092	41.04***	0.0000	0.03655	477.36***	0.0000
Velocity	1	0.00412	183.8***	0.0000	0.0066	86.22***	0.0000
Temperature × Variety	3	0.00019	8.27***	0.0003	0.00174	22.68***	0.0000
Temperature × Velocity	1	0.00028	12.63***	0.0011	0.00208	27.19***	0.0000
Variety × Velocity	3	0.00005	2.38 n.s.	0.0863	0.00076	9.88***	0.0001
Error	35	0.00002			0.00008		
Total	47						

d.f. – degree of freedom, *MSE* – mean square error, F – statistical distribution, P – test statistic probability, n.s. – not significant, *** highly significant.

T a b l e 5. Effective moisture diffusivity for each experiment

Temperature (°C)	Velocity (m s^{-1})	Pedro		Serr		Z67		K82	
		$D_{eff}\times10^7$	R^2	$D_{eff}\times10^7$	R^2	$D_{eff}\times10^7$	R^2	$D_{eff}\times10^7$	R^2
43	1	4.96	0.9690	6.06	0.9897	5.90	0.9878	9.30	0.9972
43	3	6.54	0.9886	6.91	0.9959	6.93	0.9927	9.92	0.9967
32	1	3.54	0.9879	4.53	0.9849	4.85	0.9847	6.84	0.9941
32	3	4.87	0.9855	4.56	0.9672	4.88	0.9797	8.04	0.9955

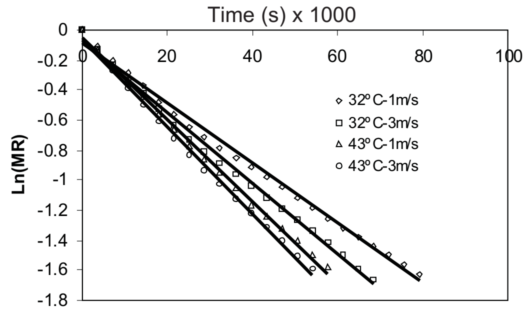

Fig. 6. ln(*MR*) versus time(s) for K82 variety.

CONCLUSIONS

1. Walnut drying occurred in the falling rate period.
2. Variety, drying air temperature and air velocity are significant factors in drying time and Page model constants of walnuts.
3. Drying air temperature is the most important factor in drying of walnuts. Higher drying air temperature resulted in a shorter drying time.
4. The Page model was the best mathematical model for describing drying kinetics of walnuts.
5. The Page model constants of k and m can be predicted as a function of air temperature, air velocity and variety properties.
6. The effective diffusivity was calculated from the data and varied from 3.54×10^{-7} to 9.92×10^{-7} m^2 s^{-1} with the variables dependence represented by an Arrhenius-type relationship.
7. Drying time can be predicted as a function of air conditions and variety properties.

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