MODELING OF CONVECTIVE DRYING KINETICS OF PISTACHIO KERNELS IN A FIXED BED DRYING SYSTEM

by

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Original scientific paper
DOI: 10.2298/TSCI110307040B

Drying kinetics of pistachio kernels with initial moisture content of 32.4% wet basis was investigated as a function of drying conditions in a fixed bed drying system. The drying experiments were carried out at different temperatures of drying air (40, 60 and 80 °C) and air velocities (0.05, 0.075, and 0.1 m/s). Several experiments were performed in terms of mass of pistachio kernels (15 g and 30 g) using a constant air velocity of 0.075 m/s. The fit quality of models was evaluated using the determination coefficient, sum square error, and root mean square error. Among the selected models, the Midilli et al model was found to be the best models for describing the drying behavior of pistachio kernels. The activation energies were calculated as 29.2 kJ/mol and effective diffusivity values were calculated between 1.38 and $4.94 \cdot 10^{-10}$ *m*^{2}*/s depending on air temperatures.*

Key words: *drying, mathematical modeling, unshelled pistachio, activation energy*

Introduction

Drying process has a meaning of water vapor to be removed from the surface of any material into the surroundings, which is a complicated process consisting of simultaneous heat and mass transfer between the material surface and its surrounding [1, 2]. This complicated process depends on different factors such as air temperature/velocity, relative humidity of air, physical nature and initial moisture content of drying material [3]. The main heat transfer occurs as a result of heat of evaporation between the solid matter and the drying air, at the same time, though heat transfer also occurs between systems and surroundings [4]. Principally, high temperature air with low relative humidity is preferable for rapid drying process. On the other hand, the higher temperature of air is limited by the quality and application of the product [5].Therefore safe and controllable drying methods are required.

Drying kinetics is important in the analysis of drying process. Many thermo-physical properties and transport properties that are usually integrated in a drying model can be determined from the analysis of drying kinetics [6]. In recent studies, modeling of the drying kinetics of foods has been widely investigated, which focuses on the products such as coconut [7], strawberry [8], olive pomace [9], and seeded grape [10]. Also, some of these are about on the drying process of pistachios. For example, Midilli [11] studied experimentally drying behavior and

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conditions in a solar drying system, Kashaninejad *et al*. [12] studied thin-layer drying characteristics and modeling of pistachio nuts growing in Iran. Johnson *et al*. [13] studied about the simulation of commercial pistachio drying in a forced-air oven. Midilli and Kucuk [14] performed a mathematical model of thin layer drying of pistachio by using solar energy.

The pistachio production is mostly seen in countries having a climate of warm arid. Iran, United States, Turkey, Italy, and Syria are the leaders of pistachio production. Turkey is one of the most pistachio-producer countries with 120113 tons, which, in other words, makes Turkey the third largest pistachio-producer after Iran (192269 tons) and USA (126100 tons) [15]. A large percentage of pistachios are marketed as the shelled and unshelled pistachios. Shelled pistachios are used for snack food as roasted and unshelled pistachios are used for both snack food and dessert suppliers. The correctly processing after harvest is very significant on pistachio quality that determines earnings and marketability.

Materials and methods

Fresh pistachios were harvested in an orchard in Siirt province, Turkey. Pistachio cultivars of Siirt are the main pistachio variety in Siirt. Siirt is located at 37º 56 N and 41° 57 E and 895 m above sea level in the southern east of the Anatolia. The harvested pistachios were peeled and separated from the outer shells for the experiments. Average length, equivalent diameter and mass of pistachio kernels were measured as 1.8 cm ± 0.1 cm, 0.9 ± 0.05 cm, and 0.8 ± 0.2 g, respectively. The initial moisture content of pistachio kernels (PKs) was determined as follows: 30 g of sample was put down in the high-temperature oven, and kept the temperature inside at 105 °C during the time taking six hours. So, the initial moisture content of PKs sample was obtained as 32.4% (w. b.).

Experimental set-up

The experimental set-up was designed and fabricated in the laboratory in order to dry some matters. The experimental set up is presented as a 3-D schematic view in fig. 1. The system consists of a steel frame, an air compressor, two air filters, a flow meter, a thermometer and hygrometer, an electrical heater, a thermometer, an insulated pipe, a glass tube, a drying cap, a digital scale, a heater controller, and a notebook. Air supplied from the compressor after filtered by inlet and outlet filters with, respectively, was heated by electrical heater and fed to the drying cap having an inner diameter of 50 mm. The temperature of the heating medium was controlled by using a heater controller having an accuracy of ±0.5 °C connected to the electrical heater. Before the drying experiments, air flow rate was measured by the method given in fig. 2. As seen in fig. 2, the apparatus consists of inlet and outlet flow meters (accuracy: ±0.5%). The inlet flow meter was calibrated with respect to outlet flow meter. The desired flow rate values adjusted by

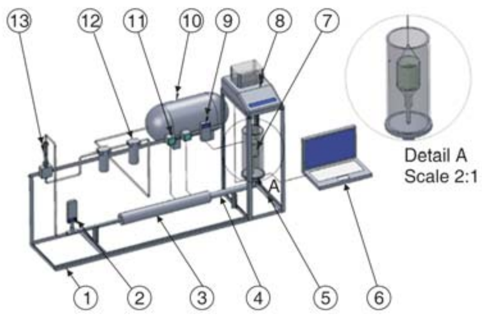

Figure 1. The schematic view of experimental set-up;
(1) – Steel frame, (2) – Thermometer and hygrometer, (3) – Electrical heater, (4) – Insulated pipe, (5) – Glass tube, (6) – Notebook, (7) – Drying cap, (8) –Digital scale, (9) – Thermometer, (10) – Air compressor, (11) – Heater controller, (12) – Air filters, (13) – Flow meter

the outlet flow meter were converted to air velocity. Air velocities were kept constant at the values of 0.05, 0.075, and 0.1 m/s for all temperatures of 40, 60, and 80 ºC. Drying experiments was started when drying conditions were achieved constant air temperatures. After, the PKs were placed inside the drying cap and the measurements were recorded. A mass of 30 g of PKs was found to be corresponded to a bed height of about 22 mm in the drying cap. Temperatures at inlet and outlet of the bed, relative humidity of air, air velocity and mass loss of PKs were measured at the end of every 20 minutes. Mass loss was measured by the digital scale having an accuracy of 0.001 g. The samples were dried until no distinct difference between subsequent readings was observed. Each experiment was lasted about 7 hours. The drying data from the different drying runs were expressed in terms of the moisture ratio (*MR*) *vs.* drying time and the drying rate *vs.* drying time.

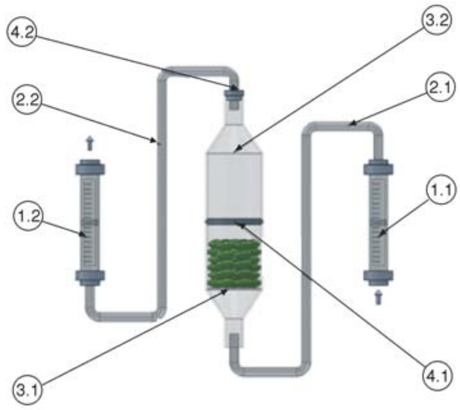

Figure 2. The schematic view of experimental set-up for calibration of air flow rate;
(1.1) – Inlet flow meter, (1.2) – Outlet flow meter, (2.1) – Inlet connecting pipe (2.2) – Outlet connecting pipe, (3.1) – Inlet drying cap, (3.2) – Outlet drying cap, (4.1) – Seal element for connecting inlet and outlet drying caps, (4.2) – Seal element for providing no leakage in between outlet drying cap and outlet connecting pipe

Mathematical modeling of drying curves

Simplified drying models in the literature have been used to describe the drying kinetics of PKs. The moisture ratio (*MR*) of PKs is expressed by eq. 1. In this study, the mass change data obtained in drying experiments were converted into the *MR*. The *MR* was simplified to M_t/M_0 since the value of equilibrium moisture content (M_e) is relatively small as compared to M_t and M_0 [13, 16].

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

The correlation coefficient (R^2) is one of the primary criteria to select the fit quality of the models [14], which has a range from 0 to 1. In addition to R^2, sum square error (*SSE*), and root mean square error (*RMSE*) are used to determine the quality of the fit [9, 17]. The higher R^2 values and the lower *RMSE* values provide a better quality of fit [8, 9, 18, and 19].

The *SSE* and *RMSE* can be calculated as follows:

$$SSE = \frac{1}{N}\sum_{i=1}^{N}(MR_{ei} - MR_{pi})^2 \tag{2}$$

$$RMSE = \sqrt{\frac{1}{N}\sum_{i=1}^{N}(MR_{ei} - MR_{pi})^2} \tag{3}$$

Analysis of drying rates

The drying rates of PKs were calculated by the following equation:

$$DR = \frac{M_{t+dt} - M_t}{dt} \tag{4}$$

Calculation of the effective moisture diffusivity

The effective moisture diffusivity is an important mass transport property needed in the modeling various drying food process. The most widely investigated theoretical model in drying of different products in the falling period is given by the solution of Fick's 2nd law of diffusion [20]. Fick's 2nd law of diffusion describes the rate of accumulation (or depletion) of concentration within the volume as proportional to the local curvature of the concentration gradient:

$$\frac{M_t - M_e}{M_0 - M_e} = \frac{8}{\pi^2} \sum_{n=0}^{\infty} \frac{1}{(2n+1)^2} \exp\left(-\frac{(2n+1)^2 \pi^2 D_e t}{4L^2} \right) \tag{5}$$

$$\ln MR = \ln\left[\frac{8}{\pi^2}\right] - \left[\frac{\pi^2 D_e}{4L^2}\right] t \tag{6}$$

The effective moisture diffusivity was calculated using the method of slopes obtained from fitting experimental data of drying model. Diffusion coefficients are typically determined by plotting experimental drying data in terms of ln *MR vs.* time (as given in eq. 6) [21]. From eq. (6), a plot of ln *MR vs.* time gives a straight line with a slope can be calculated as:

$$\text{Slope} = -\frac{\pi^2 D_e}{4L^2} \tag{7}$$

Estimation of activation energies

The activation energy can be determined from the slope of the Arrhenius plot of lnD *vs.* $1/T_a$:

$$D_e = D_0 \exp\left(-\frac{E_a}{R_g T_a} \right) \tag{8}$$

From eq. 7, a plot of ln *D vs.* $1/T_a$ gives a straight line whose slope *K* to be found as:

$$K = \frac{E_a}{R_g} \tag{9}$$

Results and discussion

Analysis of drying curves

The effect of different air temperatures at constant air velocity on drying of PK is shown in fig. 3. The increase in drying temperature at constant air velocity speeds up the drying process therefore it shortens the drying time. The drying air temperatures have a significant effect on the drying kinetics of unshelled pistachios. At the drying temperatures of 40, 60 and 80 °C, the drying times to reach of a moisture content of 5% (d. b.) were about 220, 300, and 360 minutes, respectively.

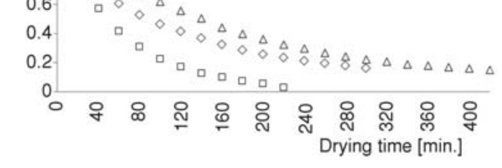

Figure 3. *MR vs.* drying time of PKs at different temperatures (mass of sample = 30 g)

Figure 4 shows the effect of air velocities of 0.05, 0.075, and 0.1 m/s on *MR* at constant temperatures of 60 ºC. The air velocity of drying air does not have effect on removing moisture of PKs as much as drying air temperature. An increase in drying air velocity of PKs from 0.075 to 0.1 m/s has a relatively small effect on decreasing drying time. The effect of bed height on drying of PKs is presented in fig. 5 at constant temperature of 60 ºC and a constant drying air velocity of 0.075 m/s. It can be seen in the

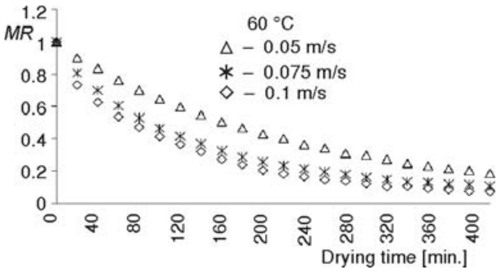

Figure 4. ***MR vs.*** **drying time of PKs at different air velocities (mass of sample = 30 g)**

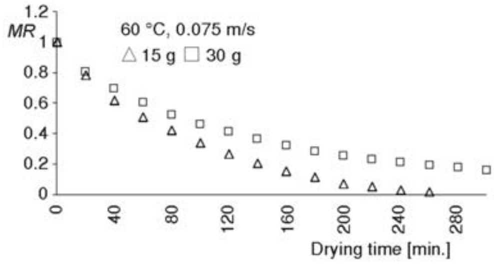

Figure 5. ***MR vs.*** **drying time of PKs at different masses**

figure that the mass of PKs in 15 g affects the drying time much more than the mass in 30 g. The mass of 15 g unshelled pistachios requires shorter processing time.

Modeling of drying curves

The data of moisture content obtained from the drying experiments were analyzed using MATLAB software to seven mathematical models listed in tab. 1. The models were fitted to observed data, and the comparison was performed using goodness-of statistical parameters. The values of R^2, *SSE,* and *RMSE* with coefficients for different air temperatures (40 °C, 60 °C, and 80 °C) are presented in tab. 2. As it can be seen, the R^2, *SSE,* and *RMSE* values for all runs range from 0.9751 to 0.9998, 0.0001703 to 0.03227 and 0.00008387 to 0.0392, respectively. The highest R^2 values and the lowest *RMSE* values in all cases were found by Midilli and Kucuk model. The R^2 values of the Midilli and Kucuk model vary between 0.9997 and 0.9998. Also, the *RMSE* values vary between 0.0001703 and 0.0004206. Figure 6 shows the graph of variation of experimental and predicted *MR* by Midilli and Kucuk model with drying time at different temperatures. As can be seen in fig. 6, Midilli and Kucuk model has a good agreement with the experimental data for different temperatures.

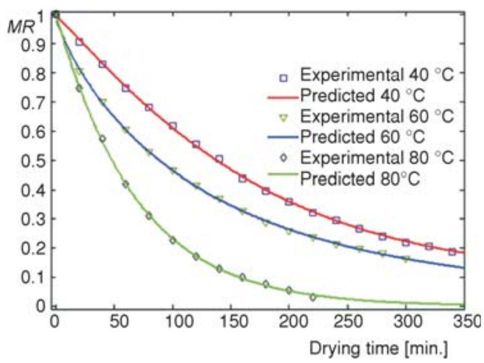

Figure 6. Variation of the experimental and predicted *MR* with drying time by Midilli and Kucuk model

Table 1. Mathematical models applied to drying curves

No.	Model name	Model	References
1	Newton	$MR = \exp(-kt)$	[17]
2	Page	$MR = \exp(-kt^n)$	[22]
3	Henderson and Pabis	$MR = a\exp(-kt)$	[23]
4	Logarithmic	$MR = a\exp(-kt) + c$	[24]
5	Two term	$MR = a\exp(-k_0t) + b\exp(-k_1t)$	[25]
6	Wang and Singh	$MR = 1 + at + bt^2$	[26]
7	Midilli and Kucuk	$MR = a\exp(-kt^n) + bt$	[14]

Table 2. Statical results of seven mathematical models

Model	Constants	T °C	R^2	*SSE*	*RMSE*
Newton	k = 0.004962	40	0.9983	0.002566	0.01105
	k = 0.006795	60	0.9751	0.03227	0.0392
	k = 0.01453	80	0.9997	0.0003493	0.005635
Page	k = 0.005109, n = 0.9945	40	0.9983	0.002549	0.01129
	k = 0.02046, n = 0.7856	60	0.9994	0.0008029	0.006336
	k = 0.01363, n = 1.014	80	0.9997	0.000291	0.005394
Henderson and Pabis	a = 1.003, k = 0.004977	40	0.9983	0.002547	0.01128
	a = 0.9363, k = 0.006519	60	0.989	0.01004	0.02677
	a = 1.003, k = 0.01458	80	0.9997	0.0003379	0.005813
Logarithmic	a = 0.9762, c = 0.0367, k = 0.005433	40	0.9989	0.001654	0.009331
	a = 0.8549, c = 0.119, k = 0.009109	60	0.9978	0.002045	0.01254
	a = 1.006, c = –0.004932, k = 0.01438	80	0.9997	0.0003069	0.00584
Two term	a = 1.01, b = 3.089e-005, k_0 = 0.005107, k_1 = – 0.01671	40	0.9994	0.006826	0.00008387
	a = 0.2349, b = 0.7604, k_0 = 0.02739, k_1 = 0.005261	60	0.9994	0.0005483	0.006759
	a = –0.009284, b = 1.009, k_0 = 5.407, k_1 = 0.01467	80	0.9997	0.0003117	0.0062242
Wang and Singh	a = –0.004309, b = 5.568e-006	40	0.9984	0.002291	0.0107
	a = –0.006366, b = 1.249e-005	60	0.9757	0.0222	0.0398
	a = –0.01064, b = 2.958e-005	80	0.9835	0.01726	0.04155
Midilli *et al.*	a = 0.9931, b = 0.0001638, k = 0.00306, n = 1.112	40	0.9997	0.0004206	0.004834
	a = 0.9982, b = 5.243e-005, k = 0.001737, n = 0.8245	60	0.9998	0.0001703	0.003767
	a = 0.9995, b = –2.246e-006, k = 0.01364, n = 1.014	80	0.9997	0.00022904	0.006024

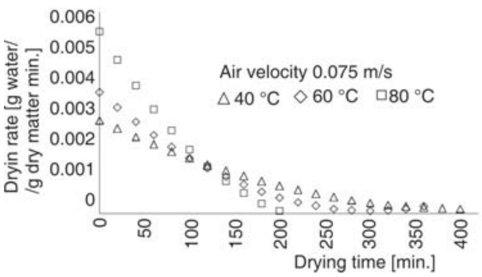

Figure 7. Drying rate *vs.* drying time of PKs at different temperatures

The drying rates *vs.* drying time of PKs were shown in fig. 7 and fig. 8 at the different temperatures. As it can be seen from figures, during the period of decreasing drying rate, the effect of moisture content continuously decrease. Towards end of the process, when the drying rate value is about 0.001, which is completely controlled by internal diffusion, the air velocity has no longer any effect. Drying rate *vs.* *MR* for PKs at different temperatures is shown in fig. 9. Drying rate increases with the increase in air temperature and the highest values of drying rate were obtained during the experiments at 80 °C of the drying air. In higher temperatures, because of increasing in heat transfer potential between drying air and PKs, evaporation of water inside the unshelled pistachios accelerates.

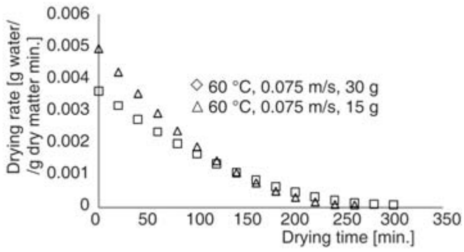

Figure 8. Drying rate *vs.* drying time of PKs at different masses/bed heights

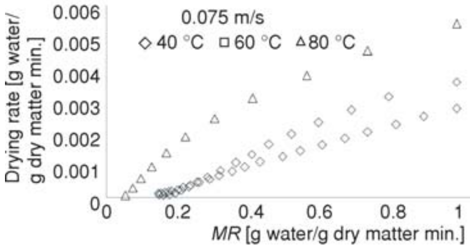

Figure 9. Drying rate *vs. MR* of PKs at different drying air temperatures

All the drying process occurred in falling rate period, indicating that the internal mass transfer has taken place by diffusion. In other words, this shows that diffusion-controlled process in which the rate of moisture removal is limited by diffusion of moisture from inside to surface of product.

In this study, the value of activation energy and effective moisture diffusivity were calculated at different drying air temperatures in constant air velocity (0.075 m/s). The value of activation energy depending on different temperatures was calculated as 29.2 kJ/mol and the effective moisture diffusivity values were calculated between $1.38\cdot10^{-10}$ and $4.94\cdot10^{-10}$ m^2/s.

Conclusions

In this study, drying kinetics of PKs were investigated in a fixed bed drying system at different temperature of drying air (40, 60, and 80 ºC) and air velocities (0.25, 0.5, and 1 m/s). The obtained results are summarized as follows:

- the initial moisture content of PKs was determined as 32.4% w. b. in the high-temperature oven at 105 ºC for 6 hours,
- increasing the temperature gradually increases the *MR* of PKs for the same period of drying time and decreases the total drying time,
- the drying rate in shorter bed height is faster than in higher bed height during the drying period,
- Among the selected models, Midilli and Kucuk model showed a good agreement with the experimental data with higher coefficient of determination value,
- the R^2, *SSE,* and *RMSE* values for Midilli and Kucuk model range from 0.9997 to 0.9998, 0.0001703 to 0.0004206, and 0.0033767 to 0.006024, respectively,
- a constant rate period was not observed in the drying of unshelled pistachios; all the drying process occurred in falling rate period, and
- the activation energy was calculated as 29.2 kJ/mol and also effective diffusivity values were calculated between $1.38\cdot10^{-10}$ and $4.94\cdot10^{-10}$ m^2/s depending on air temperatures.

Nomenclature

D_e – effective moisture diffusivity, [m^{2}s^{-1}]
D_0 – constant, [–]
E_a – activation energy, [Jmol^{-1}]
K – slope, [–]
L – half thickness of slab, [m]
M_t – moisture content at time *t*, [g water/g dry matter]
M_0 – initial moisture content, [g water/g dry matter]
M_e – equilibrium moisture content, [g water/g dry matter]
MR – moisture ratio, [–]
MR_{ei} – i^{th} experimental moisture ratio, [–]
MR_{pi} – i^{th} predicted moisture ratio, [–]
N – number of observations, [–]
R_g – universal gas constant, [kJ/ mol °K]
R^2 – coefficient of determination, [–]
RMSE – root mean square error, [–]
SSE – sum square error, [–]
T – drying temperature, [°C]
T_a – absolute temperature, [K]
t – drying time, [min.]
w. b. – wet basis, [g water/g total mass]
d. b. – dry basis, [g water/g total mass]

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Paper submitted: March 7, 2011
Paper revised: February 14, 2013
Paper accepted: March 11, 2012