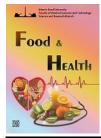
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The effects of solar drying on drying kinetics and effective moisture diffusivity of pistachio nut

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This research aimed to investigate the process of pistachios solar drying in two different conditions of the designed dryer (recycle and without recycle modes). As well, in this study, the drying kinetics curves of pistachio were drawn and its dewatering behavior was monitored by empirical models. Furthermore, the mass transfer rate (MTR) of the examined product during drying in two noted modes was determined by Fick's 2^{nd} law. Maximum, minimum, and average values of collector output air temperature were recorded 54.20, 32.30, and 42.99°C & 47.10, 30.70, and 40.03°C in air recycle and without air recycle modes, respectively. The results dedicated that, 18.75 % saving in drying time was obtained for air recycle mode compared with without air recycle mode. Also, the obtained data revealed that the drying rate of pistachio nut under recycle mode was higher than that of pistachio nut under without recycle mode, especially during the first hours of drying. Moreover, the empirical modeling showed that the Parabolic and Diamante et al. models were found to be the most suitable for describing drying curve of the thin layer solar drying process of whole pistachio nut under recycle and without recycle modes, respectively. In addition, the highest value of effective moisture diffusivity was observed in solar dried pistachio nut under air recycle mode with the D_{eff} value of $2.26 \times 10^{-6} \text{ m}^2/\text{h}$ (R²=0.9265).

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1. Introduction

Pistachio nut (*Pistacia vera L.*) is one of the main products of Iran, USA, and Turkey. According to the food and agricultural organization (FAO), production of pistachio nut in 2018 was reported 551307 MT, which Iran is dedicated ~38% of the global production of pistachio nut this year. According to the official of the FAO, Iran is the largest producer of pistachio nuts in the world. The main parts of pistachio production are exported, economically, it is an important source of non-oil foreign currency income (1). Complete processing line of this valuable product is including peeling, washing, separation, drying, sorting, packaging and storage. Drying is one of the critical steps in the processing of dehydrated products and it is energy-intensive and has a negative environmental impact due to the fact that most of the

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thermal energy needed is obtained by combusting fossil fuels (2). Thus, selecting the appropriate method of drying can be further savings in energy consumption, causing improved color and appearance attributes of the product. Pistachio nuts are commonly preserved by open-air sun drying or industrial drying methods after harvest. However open-air sun drying has some problems, including the contamination with dust, soil, sand particles, and insects and long drying times (3). Also, high initial cost, accumulation of greenhouse gases in the atmosphere, and energy consumption are some problems associated with the industrial drying method, so, this method is not attention for gardeners and pistachio recording terminals. To overcome previous problems, the drying process can be replaced with solar drying. Solar energy is an important alternative energy source and preferred to other energy sources because it is plentiful, interminable, and non-pollutant. Also,

it is renewable energy, cheap, and environmentally friendly (4). According to the renewable energy organization of Iran, (REOI) utilization of solar energy as a reliable energy source to dry foods has great potential, as, the daily average solar radiation on a horizontal plane in Iran is 4.5-5 kW/[m².day] with 300 sunny days (5). Recently, there have been many studies on the drying characteristic of pistachio nuts using different drying techniques. For example, Mokhtarian et al (6) studied the effects of solar drying along with air recycling system on physicochemical and sensory properties of dehydrated pistachio nuts. The results indicated that the pistachio dehydrated with conventional solar drying along with air recycling had the lowest shrinkage and the highest drying rate, shell splitting, bulk density, and kernel density in comparison with conventional solar drying. As well, the pistachio dried with conventional solar drying along with air recycling (had the best sweetness and roasting flavor) in comparison with those dried with conventional solar drying because of the higher drying rate. In another research, Mokhtarian et al (7) studied the energy and exergy analysis of whole pistachio nut under solar drying conditions (with and without air recycling, respectively named with Greek numbers of I and II). The drying air of Method I obtained ~55% more enthalpy from its solar collectors and transferred ~35% more heating energy to the product than the one in Method II. Consequently, its thermal and pickup efficiencies became, respectively, 40 and 80% more than Method II. Overall, the pistachio dried with Method I used much less energy than those dried with Method II and had a higher quality than those dried with commercial driers due to drying temperature $<50^{\circ}$ C. Tavakolipour (8) found that the time required for drying of Kerman variety pistachio nuts in convective hot air dryer from the initial moisture content of 47% (wet basis) to the final moisture content of 4.5% (wet basis) as a monolayer at 50°C and a velocity of 1 m/s took approximately 5 h. Kouchakzadeh (9) examined the effect of solar and acoustic energy on the pistachio drying process. The results revealed that the application of acoustic energy-assisted solar energy resulted in lower power consumption, greater economic saving, and reduce drying time to 4 hours. Tavakolipour and Mokhtarian (10) claimed that the Modified Page model was found to be the best model for describing the drying behavior of the pistachio cultivar with high R²-values (0.99). Thin-layer drying characteristics and modeling of pistachio nuts in an air recirculating dryer unit were investigated by Kashaninejad et al (11). They found that the drying process of pistachio nut took about 11 hours at 55°C, RH=5%, and air velocity of 1.5 m/s (the initial moisture content was determined in the range of 36-37 % w.b). Gazor and Minaei (12) investigated the effect of drying conditions (air temperature and air velocity) on drying time and quality parameters of pistachio (Pistacia vera L.) in laboratory drier and they concluded that the drying air temperature was the main factor in controlling the rate of drying. The results of these researchers show that, when air velocity increased from 1.5 to 2.5 m/s, drying time reduced an average of 9.43% for Kaleghochi and Fandoghi varieties whereas increasing the temperature from 60 to 90°C decreased drying time by about 35 to 40%. Midilli and Kucuk (13) studied mathematical modeling of thin-layer drying of pistachio nut by using solar energy and deduced that the logarithmic model could sufficiently describe thin layer forced solar drying of shelled and unshelled pistachio, while the two-term model could define thin layer natural solar drying of these products. The aim of this research was to investigate the process of pistachios solar drying in two different conditions of the designed dryer (recycle and without recycle modes). As well, in this study, the drying kinetics curves of pistachio were drawn and its dewatering behavior was monitored by empirical models. In addition, the mass transfer rate (MTR) of the examined product during drying in two noted modes was determined by Fick's 2nd law.

2. Materials and methods

2.1. Raw materials preparation

Fresh Kalle-Ghuchi variety whole Pistachio (*Pistacia vera L.*) as raw material was purchased daily from one of the gardens of Keyzur village, Sabzevar county, Khorasan Razavi province in Iran during the summer season in the 2014 year. Before drying, Pistachio was manually peeled and cleaned to remove all foreign matters as well as immature and broken nuts. To prevent drying of surface moisture, after peeling it was immediately placed in a plastic container and sealed. Then, Pistachios were stored until the start of the experiments in a refrigerator at 3°C.

2.2. Drying procedure

In this study for drying of pistachio nut, a designed solardried was used. The experiment was carried out in the shiny area of Sabzevar Azad University. The experimental location has latitude $36^{\circ}13$ N and longitude $57^{\circ}37$ E. Whole pistachio nuts were dried at different operational modes.

Mode (A): drying using the indirect active solar dryer with air recycles state: the air recycle was carried out when the relative humidity of the drying chamber output was less than 15% (RH \leq 15%) by the electronic controlling unit.

Mode (B): drying using the indirect active solar dryer without air recycles state.

2.3. Data fitting

Thin-layer drying models were used for the mathematical modeling of the drying curves of the pistachio nut in various drying modes. In these models, the variation of moisture content in the falling rate drying period is proportional to the instantaneous difference between the material moisture content and the expected material moisture content when it comes into equilibrium with the drying air. Thin-layer drying theory assumed that the material layer is thin enough or the air velocity is high so that the conditions of the drying air (humidity and temperature) are kept constant throughout the material. The obtained drying curves were fitted with 19 different moisture ratio models (14). The equations of these models are given in Table 1.

Table 1. Thin	layer	drying	kinetics	models.
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Model	Model name	Model equation		
<u>no.</u> 1	A abbashlo at al	$MP = ovp(\frac{1}{t}t/1 + \frac{1}{t}t)$		
-	Aghbashlo et al	$MR = exp(-kt/1+k_1t)$		
2	Diffusion of Fick's	$MR = a \exp(-c(t/L^2))$		
3	Peleg	MR = 1 - (t/(a+kt))		
4	Page	$MR = exp(-kt^n)$		
5	Modified page II	$MR = a \exp(-c(t/L^2)^n)$		
6	Thomson	$t = a \ln (MR) + b (\ln (MR))^2$		
7	Diffusion approximation	$MR = a \exp(-kt) + (1-a) \exp(-kbt)$		
8	Two-term	$MR = a \exp(-k_0 t) + b \exp(-k_1 t)$		
9	Two-term exponential	$MR = a \exp(-kt) + (1-a) \exp(-kat)$		
10	Diamante et al	$\ln (-\ln MR) = a + b (\ln t) + c (\ln t)^2$		
11	Parabolic	$\mathbf{MR} = \mathbf{a} + \mathbf{bt} + \mathbf{ct}^2$		
12	Silva et alii	MR=exp(-at-bt ^{0.5})		
13	Logarithmic	$MR = a \exp(-kt) + C$		
14	Midli-kucuk	$MR = a \exp(-kt^n) + bt$		
15	Newton	MR = exp(-kt)		
16	Wang and Singh	$\mathbf{MR} = 1 + \mathbf{at} + \mathbf{bt}^2$		
17	Verma et al.	$MR = a \exp(-kt) + (1-a) \exp(-gt)$		
18	Henderson and Pabis	$MR = a \exp(-kt)$		
19	Modified Henderson and Pabis	MR=a exp(-kt) +b exp(-gt) +c exp(- ht)		

The coefficient of determination R^2 was one of the main criteria for selecting the best equation. In addition to the coefficient of determination, the goodness of fit was determined by various statistical parameters such as reduced chi square (χ^2), mean relative deviation modulus P (%), and root mean square error RMSE. For quality fit, R^2 value should be higher and χ^2 , P (%), and RMSE values should be lower (10). The above parameters can be calculated as follows:

$$R^{2} = 1 - \left[\frac{\sum_{i=1}^{N} (MR_{p,i} - MR_{e,i})^{2}}{\sum_{i=1}^{N} (\overline{MR}_{p,i} - MR_{p,i})^{2}}\right]$$
(2)

$$RMSE = \left(\frac{1}{N}\sum_{i=1}^{N} (MR_{p,i} - MR_{e,i})^2\right)^{0.5}$$
(3)

$$X^{2} = \frac{\sum_{i=1}^{N} (MR_{e,i} - MR_{p,i})^{2}}{N-z}$$
(4)

$$P(\%) = \frac{100}{N} \sum_{i=1}^{n} \left| MR_{p,i} - MR_{e,i} \right|$$
(5)

Which $MR_{e,i}$ is ith experimental moisture ratio, $MR_{p,i}$ is ith predicted moisture ratio, $\overline{MR}_{p,i}$ is average predicted moisture ratio, N is the number of observations and z is the number of model's constants. Also, mathematical modeling was carried out by Sigma Plot software version 12.

2.4. Determination of effective moisture diffusion

Drying characteristics of agricultural and food products in the falling rate period (FRP) can be described by using Fick's second law in diffusion (15, 16). The Fick's second law solution for spherical geometry given by Crank (1975) is as follows (17):

$$\frac{\partial X}{\partial t} = \frac{1}{r} \frac{\partial}{\partial r} \left(D_{eff} r \frac{\partial X}{\partial r} \right) \tag{6}$$

In this equation, X is the moisture content (d.b.), r is the pistachio nut radius (m), D_{eff} is the effective moisture diffusivity (m²/s), t is the drying time (s). For the solution of equation 6, the following assumptions were considered: Mass transfer from the center of the sphere to its outer surface in the falling rate period occurred as diffusion form and pistachio nut shrinkage was negligible. Temperature, relative humidity, and pressure of ambient air during the drying process were assumed constant. Initial and boundary conditions for sphere in radius can be written as follows:

(1) t = 0, $X = X_0$, 0 < x < r(2) t > 0, $X_t = X$, x = 0 and x = r(3) t >> 0, $\partial X / \partial t = 0$, x = 0

Where, r is the pistachio nut radius in m, X_t is the moisture content of the pistachio samples at any time in db, x is radial coordinate in m (in the center x=0 and on the outer surface of sphere x= r). In one dimensional (1-D) thin layer drying of pistachio with uniform distribution of moisture content in the nuts and negligible resistance at the outer surface of sphere, moisture diffusivity can be calculated using simplification of Eq. (6) as follows (17):

$$X_{t} = X_{e} + \frac{6}{\pi^{2}} (X_{0} - X_{e}) \sum_{n=1}^{\infty} \frac{1}{n^{2}} \exp\left(\frac{-D_{eff} n^{2} \pi^{2} t}{r^{2}}\right)$$
(7)

Where X_0 is the initial moisture content (d.b.), X_e is the equilibrium moisture content (d.b.), X_t is the moisture content at any time (d.b.) and n is the number of considered terms (1, 2, 3, . . .). Dimensionless form of Eq. (7) can be written as follows:

$$MR = \frac{X_t - X_e}{X_0 - X_e} = \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} \exp\left(\frac{-D_{eff} n^2 \pi^2 t}{r^2}\right)$$
(8)

Where MR is the moisture ratio (dimensionless). For a long drying period, the first term of Eq. (8) can be considered for moisture diffusivity (D_{eff}) calculation. Therefore, with substituting n=1 and taking logarithm from both sides of the above equation:

$$\ln MR = \ln \frac{X_t - X_e}{X_0 - X_e} = \ln \frac{6}{\pi^2} - \frac{D_{eff} \pi^2}{r^2} t$$
(9)

Therefore, effective diffusivity (D_{eff}) was obtained by plotting lnMR versus time by the equation slope (α) as following:

$$\alpha = \frac{D_{eff}\pi^2}{r^2} \tag{10}$$

Thus, with simplification:

$$\ln M R = \ln \frac{6}{\pi^2} - \alpha t \tag{11}$$

3. Results and discussion

3.1. Drying kinetics

The variation of moisture content and drying rate of whole pistachio nut in the various drying modes are presented in Fig. 1 (a,b).

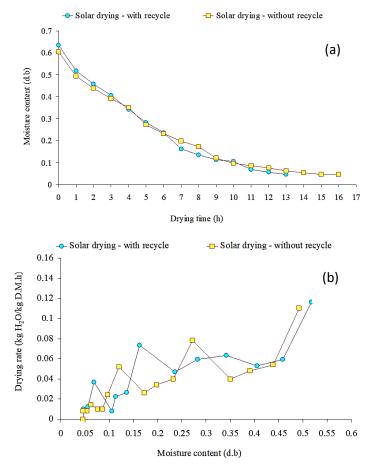


Fig. 1. Influence of the different drying methods on the drying kinetics (a) and drying rate (b) curves of whole pistachio nut.

As can be seen, in all drying modes, the trend of moisture content variation against drying time is descending. The results dedicated that; the final drying levels of the whole pistachio nut were obtained after 13 h in the solar drying system in the air recycle mode (RH≤15%) but took about 16 h in the without air recycle mode. An 18.75 percent saving in drying time was obtained for air recycle mode beside without the air recycle mode. Furthermore, the highest and the lowest drying rate of the whole pistachio were observed in the air recycle mode and without the air recycle mode, respectively. Tavakolipour (8) reported that the time required for drying of Kerman variety pistachio nuts as a monolayer at 50°C and a velocity of 1 m/s was about 5 h. Kouchakzadeh (9) studied the influence of solar and acoustic energy on the pistachio drying process. The results revealed that the application of acoustic energy-assisted solar energy resulted in lower power consumption, greater economic saving, and reduce drying time to 4 hours. Midilli and Kucuk (13) and Gazor and Minaei (12) were reported that the drying time of pistachio nuts 6 h at a temperature range of 40-60°C (the initial moisture content was estimated equal to 26.95 % w.b) and about 6 h at 60°C (the initial moisture content was estimated in the range of 36.30-38.27 %w.b), respectively. Kashaninejad et al (11) reported that the drying process of pistachio took about 11 hours at 55°C (the initial moisture content was determined in the range of 36-37 %w.b).

3.2. Data fitting of drying curves

The results of drying curves modeling of solar-dried whole pistachio nut under different modes of air recycle and without recycle are present in Tables 2 and 3. Drying curves modeling were evaluated based on R², P (%), χ^2 and RMSE. In all fitted models, R² value was estimated greater than 0.9755. The results show that, R^2 , P (%), χ^2 and RMSE ranged between 0.9755-0.9973, 0.8385-4.443, 0.000174-0.003435 and 0.01199 to 0.04873, respectively. The fitting results illustrated that Parabolic and Diamante and co-worker models were introduced as a good model for describing the drying curve of the thin layer solar drying process of whole pistachio nut under recycle and without recycle modes, respectively. Several models have been proposed for different pistachio varieties for example modified page model for Abasali cultivar (10), page and two-term models for drying of Ohadi cultivar (11, 18), page model for Kerman cultivar in convection hot air drying (16), page model for Khany and Abasali cultivars in microwave-convective drying (19). Moisture ratio variation curve of solar dried whole pistachio nut under two modes of air recycle and without recycle vs. drying time for the experimental data and predicted by the best-fitted models are presented in Fig. 2.

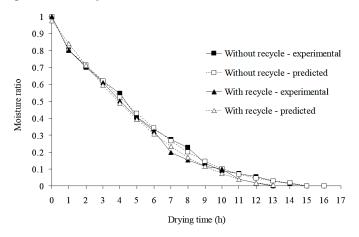


Fig. 2. Variation of experimental and predicted moisture ratios vs. drying time for whole pistachio nut in different drying modes by the best drying kinetics model: ▲, Parabolic; ■.

As can be seen, the selected models were able to predict the experimental data with a very low error (e.i. high correlation) so that, predicted values were very close to the theoretical values and their curves approximately were coincide.

3.3. Effective moisture diffusivity

The effective moisture diffusivity of solar dried whole

No.	Model coefficients*	\mathbb{R}^2	P (%)	RMSE	χ^2
1	$k = 0.1494; k_1 = -0.0421$	0.9953	1.686	0.02125	0.000526
2	a = 1.0374; c = 0.0674; L = -0.5618	0.9773	4.443	0.04684	0.002793
3	k = 0.5545; a = 5.4233	0.9937	2.0532	0.02468	0.000710
4	k = 0.1308; n = 1.260	0.9894	2.675	0.03198	0.001193
5	n = 1.260; c = 0.1401; L = 1.0278	0.9894	2.6751	0.03190	0.001302
6	a = -0.0772; b = -0.0196	0.9804	2.777	0.04281	0.002138
7	k = 0.0866; a = -114.2; b = 1.0080	0.9952	1.7666	0.02140	0.000587
8	k = 0.2237; a = 1.0872; b = -0.0872; h = 4546.5	0.9797	3.923	0.0443	0.002748
9	k = 0.2824; a = 1.7661	0.9887	2.851	0.0330	0.001271
10	a = -1.495; b = 0.4018; c = 0.2932	0.9951	0.8454	0.0128	0.000211
11	a = 0.9768; b = -0.1429; c = 0.0052	0.9962	1.525	0.0192	0.000469
12	a = 0.2644; b = -0.1332	0.9834	3.477	0.0400	0.001873
13	k = 0.1378; a = 1.2140; c = -0.2209	0.9950	1.860	0.02203	0.000617
14	k = -0.0330; n = 1.254; a = 0.9714; b = -0.1693	0.9960	1.553	0.0196	0.000538
15	k = 0.2064	0.9755	4.382	0.04873	0.002557
16	a = -0.1498; b = 0.0056	0.9954	1.542	0.0211	0.000521
17	k = 0.2064; a = 0.1277; g = 0.2064	0.9755	4.3821	0.04873	0.003022
18	k = 0.2138; a = 1.0374	0.9773	4.443	0.04684	0.002560
19	k = 145.5; a = -0.0461; b = -0.0411; c = 1.0872; g = 1623.5; h = 0.2237	0.9797	3.9233	0.04430	0.003435

Table 2. Values of statistical parameters and contact coefficients found for the selected models for thin-layer drying of whole pistachio nut in recycle mode drying (RH<15%).

Table 3. Values of statistical parameters and contact coefficients found for the selected models for thin-layer drying of whole pistachio nut without recycle mode drying.

No.	Model coefficients*	\mathbf{R}^2	P (%)	RMSE	χ^2
1	$k = 0.1448; k_1 = -0.0349$	0.9955	1.408	0.0206	0.000482
2	a = 1.0325; c = 0.1172; L = 0.7654	0.9807	3.941	0.04265	0.002209
3	k = 0.6221; a = 5.3502	0.9919	2.069	0.02769	0.000869
4	k = 0.1280; n = 1.2244	0.9901	2.513	0.03053	0.001056
5	n = 1.2244; c = 0.6150; L = 1.8979	0.9901	2.513	0.03053	0.001132
6	a = -0.0934; b = -0.0139	0.9930	2.065	0.03487	0.001378
7	k = 0.0932; a = -62.591; b = 1.0117	0.9950	1.555	0.02165	0.000569
8	k = 0.2069; a = 1.0705; b = -0.0705; h = 36.54	0.9821	3.622	0.04105	0.002203
9	k = 0.2591; a = 1.7299	0.9899	2.708	0.03091	0.001083
10	a = -1.4960; b = 0.3991; c = 0.2646	0.9973	0.8385	0.01199	0.000174
11	a = 0.9646; b = -0.1299; c = 0.0043	0.9963	1.542	0.01859	0.000419
12	a = 0.2403; b = -0.1114	0.9852	3.316	0.03741	0.001586
13	k = 0.1446; a = 1.1346; c = -0.1410	0.9945	1.674	0.02276	0.000629
14	k = -0.0309; n = 1.2331; a = 0.953; b = -0.1517	0.9951	1.857	0.02141	0.000599
15	k = 0.1941	0.9795	3.902	0.04402	0.002058
16	a = -0.1384; b = 0.0048	0.9945	1.744	0.02279	0.000589
17	k = 0.1941; a = -1.3345; g = 0.1941	0.9795	3.902	0.04402	0.002353
18	k = 0.20; a = 1.0325	0.9807	3.941	0.04265	0.002061
19	k = 22.165; a = -0.0704; b = 0.0868; c = 0.9836; g = 0.2069; h = 0.2069	0.9821	3.622	0.04105	0.002604

Table 4. Values of effective moisture diffusivity for whole pistachio drying in different drying modes.

Drying mode	le Radius (cm) D _{eff}		Reg. equation	\mathbb{R}^2
Recycle mode (RH≤15%)	0.856	2.26×10-6	$\ln MR = 0.360 - 0.3045t$	0.9265
Without recycle mode	0.856	2.10×10 ⁻⁶	$\ln MR = 0.3656 - 0.2827t$	0.9510

pistachio in various processing modes is produced in Table 4. The R² values of the regression equation for air recycle and without recycle modes were 0.9265 and 0.9510, respectively, that it is indicating the high accuracy of the regression equations. The highest value of effective moisture diffusivity was observed in solar dried pistachio nut under air recycle mode $(2.26 \times 10^{-6} \text{ m}^2/\text{h})$. Rizvi (20) stated that effective diffusivities were influenced by factors such as temperature, variety, and composition of the material among others. Also, the high effective diffusivity can be described to the low isosteric heat of sorption, hence, the restriction of moisture mobility (2). Comparison of the effective moisture diffusivity data obtained in this study with the results of Tavakolipour (16) shows that, despite the difference in the cultivar and of test conditions, it reflects closely the results of the two studies together. In this study, the effective moisture diffusivity of Damghan pistachio nut cultivar was 7.061×10^{-10} m²/s at 40°C and 2.105×10^{-9} m²/s at 70°C. Kashaninejad et al. (11) stated that the effective moisture diffusivity values of Ohadi pistachio nut cultivar were ranged between 5.42×10^{-11} to 9.29×10^{-10} m²/s.

4. Conclusions

Main conclusions remarked from the results of the present study may be listed as follows: The whole pistachio nuts in solar drying under air recycle (RH≤15%) and without air recycle modes were sufficiently dried at the ranges of drying air temperature of 42.99-54.20°C and 40.03-47.10°C during 13 and 16 h, respectively, so that, an 18.75 percent saving in drying time was obtained for air recycle mode beside without the air recycle mode. The comparison of the collector input and output air temperature showed that the solar collector was able to increase the ambient temperature by 11.47°C and 7.94°C in the various modes of air recycle and without air recycle. In order to expound the drying behavior and to develop the mathematical modeling of the whole pistachio nut, 19 models in the literature were applied to the thin layer solar drying process under recycle and without recycle modes. The results showed that the Parabolic and Diamante et al. models were found to be the most suitable for describing the drying curve of the thin layer solar drying process of whole pistachio nut under recycle and without recycle modes, respectively. Maximum and minimum values of D_{eff} were calculated under recycle and without recycle modes, respectively with the corresponding values of 2.26×10^{-6} m²/h and 2.10×10^{-6} m²/h. The results show that air-drying temperature was the most effective factor on the D_{eff} value.

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