

Drying kinetics and modeling of savory leaves under different drying conditions

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Abstract

In current work, the effects of drying temperature (40, 60, and 80°C), relative humidity (20%, 30%, and 40%) and air velocity (1.0, 1.5, and 2.0 m/s) on drying kinetics of Savory leaves were investigated in a forced convective dryer in order to optimize the drying conditions. nine most used empirical thin layer models were applied on experimental data which were selected to describe the drying behavior of the savory leaves. The selected thin layer drying models were fitted to the experimental drying curves using direct least square and the coefficients of the models were estimated under each drying air condition. The best fit was determined by using three statistical parameters: higher values of coefficient of determination (R^2) and lower sum square errors (SSE) and root mean square error (RMSE) were used, respectively. According to the statistical analysis, the Midilli et al. model was considered as the best models describing the drying curves of savory leaves by higher correlation of the coefficients and lower reduced SSE and RMSE. Effective moisture diffusivity was calculated on the basis of Fick's second law. The moisture diffusion coefficient varied between 6.76143×10^{-12} and 1.57018×10^{-10} m²/s for range of temperatures considered in different air dryer conditions and corresponding activation energy varied between 42.07 and 44.74189 kJ/mol.

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Keywords

Mathematical modeling
Air temperature
Relative humidity
Air velocity
Savory

Introduction

Savory is an aromatic herb, which has highly significant nutritional and flavor values, so it is widely used to add a distinctive aroma and flavor to food. The leaves of Savory can be used as a spice in fresh or dried form (Loughrin and Kasperbauer, 2003). In the Mediterranean diet, savory is extensively consumed, for instance; in tomato products, salads, pizza, meat, soups and marine foods. It is commonly known that the presence of essential oils and their composition determine the specific aroma of plants and the flavor of the condiment. As a spice dried, savory is usually used in bakery products, confectionary, ice creams, vinegars and flavor products (Ozcan *et al.*, 2005). To minimize physical, biochemical, chemical and microbiological deterioration of agriculture and food products, reduction of the moisture content is necessary. Therefore reduction of moisture is the most popular method for food preservation. Thermal technique is basic process to reduce the moisture content in food products (Karimi *et al.*, 2012; Naderinezhad *et al.*, 2016). Fresh fruit and vegetables have high amount of moisture and hence perishable. Drying is one of the most important unit

operations in food processing industry that employs to decrease biochemical, chemical and microbiological deterioration of food and agricultural products due to the decrease of the moisture content to the level, which allows safe storage and preservation. Also, dried products have minimized packaging, storage and transportation costs (Zielinska and Markowski, 2010; Darvishi *et al.*, 2014; Abhay *et al.*, 2016; Fathi *et al.*, 2016). Drying is one of the most traditional operations to preserve quality of aromatic, herbal and medicinal plants (Rubinskienė *et al.*, 2015). Convective drying is the most popular method for drying of food products (Motevali *et al.*, 2013). Sun and oven drying, as primary method of processing, are frequently performed by producers of raw material, often with simple techniques in bad phytosanitary conditions (Dudaš *et al.*, 2013). Arslan and Özcan (2012) studied drying process of Savory leaves with three methods; Sun, oven (50°C), and microwave oven (700 W) in constant drying condition for every method (Arslan and Özcan, 2012). In current research the drying process was investigated in variable condition of temperature, air flow velocity and relative humidity in forced convective dryer. The use of a simulation model is a significant tool

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for estimating of drying behavior of materials and evaluation performance of drying systems. Effective empirical models are essential to design of the process, optimization, energy integration, and control of dryer. Empirical modeling in food drying process are so important but theoretical model has not provided as practical as possible to unify the calculations (Torki-Harchegani *et al.*, 2015).

The main objectives of this study were to (1) dry Savory leaves in a forced conductive dryer and investigate the effect of different conditions of air temperature, relative humidity and air flow velocity on drying behavior of the samples, (2) mathematical modeling of the drying kinetics of mass transfer during air drying process of savory leaves by fitting the nine most used empirical thin layer models to the experimental data, and (3) determine the values of effective moisture diffusivity and activation energy in different drying air conditions.

Materials and Methods

Sample preparation and drying unit

The thin-layer drying experiments were conducted by forced conductive dryer which was designed and fabricated by Taheri-Garavand. The forced conductive dryer diagram was shown in Figure 1. The heating structure was consisted of ten heating elements placed inside the dryer channel. Moreover, a simple control system was applied to control and adjust the temperature, relative humidity and velocity of air used for drying process (Taheri-Garavand *et al.*, 2011a). The trays were supported by lightweight steel rods placed under the digital balance. The opening side on the right of the tunnel was employed to load or unload of material. The dryer is capable of providing any desired drying air temperature in the range of 20 to 120°C and air relative humidity in the range of 5 to 95% and air velocity in the range of 0.1 to 5.0 m/s with high accuracy. The used instruments for various measurements with their specifications are given in Table 1. The dryer was adjusted to a preset air drying condition for about 20 minutes prior to achieve the steady state.

Then, the tray holding the samples was carefully put in the dryer. The sample weight was kept constant at 50 g (±0.5 g) for all runs. During the course of the drying process, savory leaves were weighed using a digital balance connected to a computer. Three replications of each experiment were done according to a pre-set air temperature, air velocity, relative humidity and time schedule. The hot air drying was applied until the weight of samples reduced to a level corresponding to moisture content of about 0.5% d.b.

Table 1. Specifications of measurement instruments including their rated accuracy

Instrument	Model	Accuracy	Make
Digital balance	GF3000	±0.02	A&D, Japan
T-sensor	LM35	±1°C	NSC, USA
RH-sensor	SHT15	±2%	China
V-sensor	405-V1	±3%	TESTO, UK

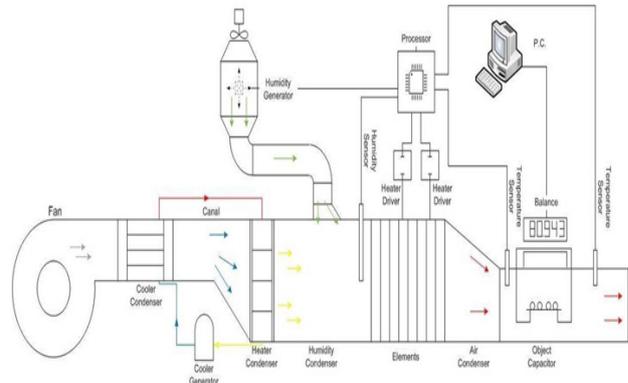


Figure 1. Scheme of pilot plan thin-layer drying equipment

The drying experiments were performed at three air temperatures of 40, 60 and 80°C and at three level of relative humidity; 20%, 30% and 40% and air flow velocity of 1, 1.5 and 2.0 m/s. The initial and final moisture contents of the savory leaves were determined at 105°C for 1 h with the oven method (Ozcan *et al.*, 2005).

Mathematical modeling of thin layer drying

The moisture ratio (MR) of savory leaves for the experimented samples was calculated according to equation (1):

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

Where MR is the dimensionless moisture content ratio; and M_d , M_0 , and M_e are moisture content at any drying time, initial and equilibrium moisture content (kg water/kg dry matter), respectively. The values of M_e are relatively low compared to those of M or M_0 , the error involved in the simplification is negligible (Aghbashlo *et al.*, 2008), as a result moisture ratio was calculated according to equation (1):

$$MR = \frac{M_d}{M_0} \tag{2}$$

The mass transfer rate that is defined as drying rate (DR) was calculated using the following equations (Xiao *et al.*, 2009):

$$DR = \frac{M_t - M_{t+\Delta t}}{\Delta t} \tag{3}$$

Where DR is drying rate (kg_{water}/kg_{dry} min); and $M_t, M_{t+\Delta t}$, are moisture content (kg water/kg dry

Table 2. Thin layer drying curve models considered and statistical results obtained from the selected models in different air condition

Model name	Model	References	R ²	SSE	RMSE
Newton	$MR = \exp(-kt)$	(Henderson, 1974)	0.91972	3.5118	0.050028
Aghbashlo <i>et al.</i>	$MR = \exp(-k_1t/1+k_2t)$	(Guarte, 1996)	0.98614	0.7875	0.02461
Page	$MR = a \exp(-kt)$	(Zhang and Litchfield, 1991)	0.97832	0.9594	0.0276412
Henderson and Pabis	$MR = \exp(-kt)$	(Aghbashlo <i>et al.</i> , 2009)	0.95588	1.9816	0.0374276
Logarithmic	$MR = a \exp(-kt) + c$	(Karathanos, 1999)	0.99204	0.5200	0.019473
Tow term	$MR = a \exp(-k_0t) + b \exp(-k_1t)$	(Yaldiz <i>et al.</i> , 2001)	0.99146	0.6437	0.020144
Wang and Singh	$MR = 1 + at + bt^2$	(Wang and Singh, 1978)	0.92998	3.766	0.05015
Modified Henderson and Pabis	$MR = a \exp(kt) + b \exp(kt) + c \exp(kt)$	(Karathanos, 1999)	0.9918	0.6989	0.02022
Midilli <i>et al.</i>	$MR = a \exp(-kt^n) + bt$	(Midilli <i>et al.</i> , 2002)	0.99394	0.4191	0.016844

matter) at t and Δt (min) drying time, respectively. Nine most used empirical thin layer models (Table 2) were selected to describe the drying behavior of the savory leaves. The selected thin layer drying models were fitted to the experimental drying curves using direct least square and the coefficients of the models were estimated under each drying air condition.

The best fit was determined by using three statistical parameters: higher values of coefficient of determination (R²) and lower sum square errors (SSE) and root mean square error (RMSE) using Equations (4-6), respectively. The statistical analyses were conducted using Matlab software (The MathWorks Inc., Natick, USA).

$$R^2 = 1 - \frac{\sum_{i=1}^N (MR_{pre,i} - MR_{exp,i})^2}{\sum_{i=1}^N (MR_{pre,i} - MR_{exp,i})^2} \tag{4}$$

$$SSE = \frac{\sum_{i=1}^N (MR_{exp,i} - MR_{pre,i})^2}{N} \tag{5}$$

$$RMSE = \left[\frac{1}{N} \sum_{i=1}^N (MR_{exp,i} - MR_{pre,i})^2 \right]^{1/2} \tag{6}$$

Where MR_{pre,i} is the *i*th predicted moisture ratio, MR_{exp,i} is the *i*th experimental moisture ratio, MR is the average of all experimental moisture ratios, N is the number of observations and n is the number of constants.

Calculation of effective moisture diffusivity and activation energy

The main mechanism of moisture removing in drying process of food and agriculture products is liquid and/or vapor diffusion. For a simple analysis of the change in moisture concentration one dimensional diffusion is considered and Fick's second law of diffusion is used in thin layer drying process. The simplified Fick's second law equation for diffusion was applied to estimate the effective diffusion coefficient of the Savory leaves during of drying process. Analytical solution of Fick's second law is represented in the equation (7), considering a constant moisture diffusivity, infinite slab geometry

and uniform initial moisture distribution (Das *et al.*, 2009).

$$MR = \frac{M}{M_0} = \frac{8}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{(2n-1)^2} \exp\left(-\frac{(2n-1)^2 \pi^2 D t}{4L^2}\right) \tag{7}$$

where M₀ is the initial moisture content (kg water/kg dry solid), MR is moisture ratio, M is the moisture content at any time (kg water/kg dry mater), n = 1, 2, 3, ... the number of terms taken into consideration, t is the time of drying in seconds, D is effective moisture diffusivity in m²/s and L is the thickness of slice (m).

For long drying periods, by neglecting the higher order terms in the above equation, it can be simplified to only the first term of the series and written in logarithmic form (Doymaz, 2012), as:

$$MR = \frac{8}{\pi^2} \exp\left(-\frac{\pi^2 D t}{4L^2}\right) \tag{8}$$

The method of slopes is employed to calculate the effective diffusion coefficient. The slopes was obtained from the linear regression of ln(MR) against the drying time according to equation (9):

$$k_0 = \frac{\pi^2 D}{4L^2} \tag{9}$$

Arrhenius' equation has been extensively employed to describe the effect of drying air temperature on the effective moisture diffusion coefficient to obtain a better agreement of the predicted curve with the experimental data. The energy of activation was calculated by using an Arrhenius type equation (Perea-Flores *et al.*, 2012):

$$D = D_0 \exp\left(-\frac{E_a}{RT_s}\right) \tag{10}$$

Where E_a is the activation energy (kJ/mol), R is universal gas constant (8.3143 kJ/mol), T_a is absolute drying air temperature (K), and D₀ is the diffusivity constant equivalent to the diffusivity at infinitely

Appendix 1. Values of the drying constant and coefficients of the best model (Midilli *et al.* model)

Temperature (°C)	Relative Humidity (%)	Velocity (m/s)	a	k(min ⁻¹)	n	b	
40	20	1	0.997	0.007182	0.9324	0.000158	
		1.5	0.9686	0.01435	0.8977	0.00002953	
		2	0.9446	0.009212	0.9187	-0.00000512	
	30	1	1.041	0.04601	0.6126	-0.002344	
		1.5	0.9937	0.02805	0.7398	-0.0001035	
		2	1.063	0.03182	0.7611	0.00009402	
	40	1	0.9738	0.01121	0.9358	0.0002188	
		1.5	1.005	0.01493	0.9365	0.0001657	
		2	1.056	0.03976	0.9042	0.0001909	
	60	20	1	0.9938	0.01933	1.207	-0.0001099
			1.5	1.009	0.03421	1.02	0.0004248
			2	1.03	0.03013	1.024	0.001133
30		1	1.058	0.03286	1.021	-0.001961	
		1.5	1.015	0.0293	1.07	0.001005	
		2	1.053	0.03628	1.014	0.0008045	
40		1	0.8789	0.01015	1.246	0.001773	
		1.5	1.021	0.03324	1.031	0.0006784	
		2	1.093	0.08947	0.7018	0.0001119	
80		20	1	0.9997	0.9152	0.9766	0.002388
			1.5	0.9907	0.05574	1.127	0.001107
			2	0.9961	0.05211	1.04	0.001149
	30	1	0.996	0.05029	1.05	0.002089	
		1.5	1.01	0.08254	0.9306	0.00171	
		2	1.02	0.09012	1.01	0.002049	
	40	1	0.945	0.07178	0.9388	0.007187	
		1.5	1.012	0.1078	0.8645	0.0007856	
		2	1.03	0.1149	0.7936	0.0004611	

high temperature (the pre-exponential factor of the Arrhenius equation) (m²/s).

The activation energy can be calculated from the slope of the Arrhenius plot, ln(D) versus 1/Ta.

From equation (10), a plot of ln(D) versus 1/Ta gives a straight slope of K1

$$K_1 = E_a / R \quad (11)$$

Results and Discussion

The drying process was stopped after no further change in sample weights. At this point, moisture content decreased from 90% to 10% (w.b.). Moisture ratio values were calculated from moisture content data and then fitted to the nine mathematical models. The results of fitting experimental data to the selected empirical models (R², RMSE and SSE) are shown in Table's 1. The best of fitting model for all drying air conditions was bolded in the mentioned tables.

The best model describing the thin layer drying kinetics of savory leaves was selected according to higher values of coefficient of determination (R²) and lower sum square errors (SSE) and root mean square error (RMSE). Based on the obtained results, Midilli *et al.* model was found the best model describing the drying process of savory leaves for all drying air conditions. Drying constant is influenced by the characteristics of the air flow conditions; temperature, velocity and relative humidity. Appendix 1 gives the drying parameters obtained using Midilli *et al.* model for the experimental data in different drying conditions. Figure 2 (a) presents the predicted moisture ratio at any particular drying temperature for different air condition (air velocity and relative humidity) by using the Midilli *et al.* model was

compared with experimental data and the results. It can be observed from the mentioned figures that the predicted moisture ratios were, generally, in line with the experimental ones. These clearly indicated that the Midilli *et al.* model could be used to describe the moisture ratio variation during the drying process of savory leaves. So this model gives a good estimation for savory drying process. The Midilli *et al.* model has also been reported by other researchers as the most suitable model to describe the drying behavior of Basil leaves (Taheri-Garavand *et al.*, 2011b), yocan slices (Shi *et al.*, 2013) and apricot kernels (Zhang *et al.*, 2016).

As can be seen from Figures 2 (a), by increasing air temperature, the heat transfer rate between thermal source (drying air) and the material (savory leaves) increases and leads to faster moisture evaporation and shorter drying. Also this Figure 2 (a) shows the variation of moisture ratio as a function of drying time. The moisture ratio of the samples reduced continually with drying time. As expected, increase in drying air temperature decreases the time required to reach any given level of moisture ratio since higher temperature results in higher heat transfer, allowing water presence in product to reach its evaporating temperature at faster rate. This increases moisture migration within the product (center to surface) due to diffusion phenomenon. This can be described by increasing temperature difference between the drying air and the samples and the resultant moisture emigration. This figure shows that a good fit can be graphically observed when using Midilli *et al.* equations.

In order to gain a deeper insight into the drying behavior of savory leaves during drying process, the variations of the drying rate with the drying time are

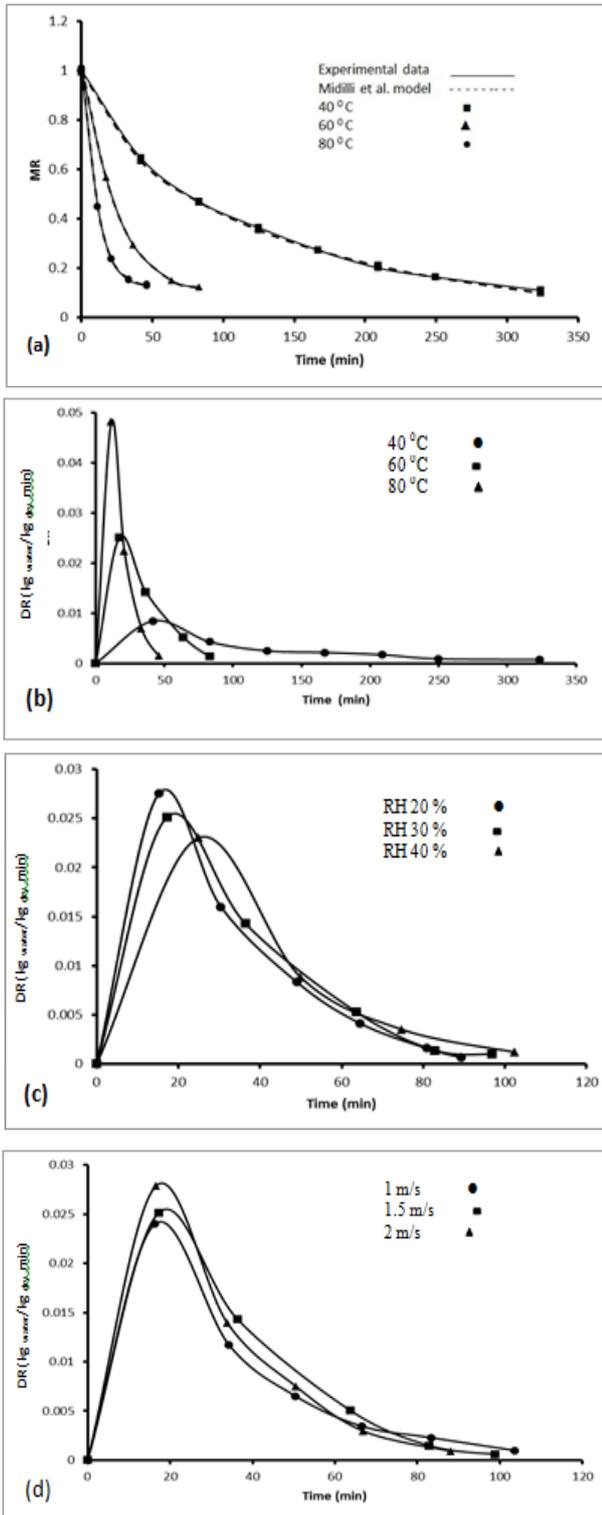


Figure 2. (a): Experimental and predicted moisture ratio by the Midilli *et al.* model versus drying time for air velocity of 1.5 m/s and relative humidity 30%. (b): Drying rate versus drying time for air velocity of 1.5 m/s and relative humidity 30%. (c): Drying rate versus drying time for air temperature 60 and air velocity of 1.5 m/s. (d): Drying rate versus drying time for air temperature 60 and relative humidity of 30%.

exhibited in Figure 2 (b-d) for various velocities, temperatures and relative humidities of the drying

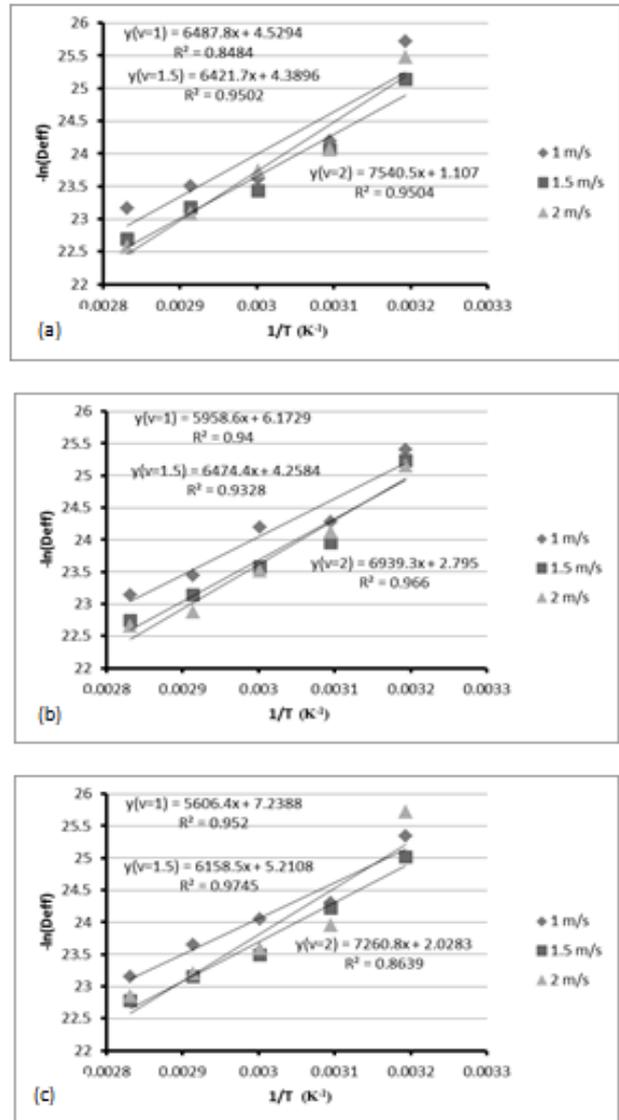


Figure 3. Relationship between effective coefficient moisture diffusivity and temperature for relative humidity of 20% (a), 30% (b) and 40% (c)

air. Due to the moisture diffusion process, the drying rate decreases with time. From Figure 2 (b), it is very obvious that higher the air temperature, higher the drying rate. For higher values of the moisture content, increasing of drying air temperature caused to increasing drying rate and next, decreasing drying time. This can be described by the increasing temperature difference between the drying air and the samples and, following, accelerating moisture migration. Other authors also reported similar results related to behavior of drying rate curves in some foods drying studies, like apricot kernels (Zhang *et al.*, 2016), peppercorns (Promvongse *et al.*, 2011), castor oil seeds (Perea-Flores *et al.*, 2012) and raw olive pomace (Koukouch *et al.*, 2015). Figure 2 (c) shows that the relative humidity has a significant influence on the variation of the drying rate with the drying time. As expected, lower relative humidity

Appendix 2. Estimated effective moisture diffusivity, activation energy and moisture diffusivity constant in different condition of air dryer

Diffusion coefficient (m ² /s)			E_a (kJ/mol)	D_0 (m ² /s)	Air velocity (m/s)	Relative humidity (%)
80°C	60°C	40°C				
8.66494×10 ⁻¹¹	5.54458×10 ⁻¹¹	6.79693×10 ⁻¹²	44.74189	1.7695×10 ⁻⁵	1	20
1.38995×10 ⁻¹⁰	8.58986×10 ⁻¹¹	1.2083×10 ⁻¹¹	43.54096	3.831×10 ⁻⁴	1.5	20
1.57018×10 ⁻¹⁰	4.93997×10 ⁻¹¹	8.6618×10 ⁻¹²	43.48715	1.28765×10 ⁻⁴	2	20
8.92166×10 ⁻¹¹	3.13039×10 ⁻¹¹	9.28141×10 ⁻¹²	43.34427	2.3563×10 ⁻⁴	1	30
1.333324×10 ⁻¹⁰	5.76324×10 ⁻¹¹	1.09596×10 ⁻¹¹	43.0121	3.09156×10 ⁻⁴	1.5	30
1.44632×10 ⁻¹⁰	6.16526×10 ⁻¹¹	1.19673×10 ⁻¹¹	42.86634	4.7092×10 ⁻⁴	2	30
8.8516×10 ⁻¹¹	3.62074×10 ⁻¹¹	9.8434×10 ⁻¹²	43.08348	2.0574×10 ⁻⁴	1	40
1.27812×10 ⁻¹⁰	6.27513×10 ⁻¹¹	1.3601×10 ⁻¹¹	42.15485	2.2245×10 ⁻⁴	1.5	40
1.19018×10 ⁻¹⁰	5.72664×10 ⁻¹¹	6.76143×10 ⁻¹²	42.07396	1.2208×10 ⁻⁴	2	40

increases drying rate change by the drying time. In other words, at low relative humidity, heat and mass transfer is high and water loss is excessive (higher difference in partial pressure of water between the air and samples). Figure 2 (d) shows drying ratio against drying time at constant air temperature and air relative humidity for air velocity 1.5, 2 and 2.5 m/s. It exposes considerable effect of air velocity on the variation of the drying rate with the drying time. As expected, higher air flow velocity intensifies drying rate change by the drying time. Thus, higher air velocity allows the higher removal of moisture from product surface.

Influence of drying air temperature, air flow velocity and relative humidity on effective moisture diffusivity and activation energy are shown in Appendix 2. For results of effective moisture diffusivity of savory leaf samples in Appendix 2, generally, temperature and air flow velocity increase have a pronounced effect on moisture diffusivity but this trend is not always straightforward. To evaluate the relationship between drying air temperature and effective moisture diffusivity we conducted a linear regression analysis as shown in Figure 3. The range of correlation coefficient is 0.8484-0.9745 which could be evaluated as a strong relationship. For instance, at 20% relative humidity, higher air velocities had a more linear trend (higher R²) than lower velocities. It could be probably due to a higher mass transfer rate and easier diffusion of heat which results in higher and more uniform moisture diffusivity values. Surprisingly at higher relative humidity (40% in our study), by increasing drying air velocity, linear trend between moisture diffusivity and temperature becomes weaker which could be explained by the fact that drying air turns to a more saturated medium in terms of diffusion of moisture and is not able to take out internal moisture easily (Usually higher air velocity should lower down saturation. might be other phenomena). Since the diffusion of air at lower velocities is lower than high

velocities, the influence of drying air with a lower velocity is stronger than higher velocity in more saturated air conditions. Activation energy was calculated and shown in Appendix 2, by increasing of air velocity and relative humidity activate energy were decreased. The moisture diffusion coefficient varied between 6.76143×10⁻¹² and 1.57018×10⁻¹⁰ m²/s for the given temperature range in different air dryer conditions. Corresponding activation energy of savory leaves varied between 42.07 and 44.74189 kJ/mol in different air dryer conditions, which was in the range of 12.7–110 kJ/mol for most food materials (Xiao *et al.*, 2012). Dai *et al.* (2015) reported this value as 31.4 kJ/mol for apricot samples treated by continuous dehumidification. Bia *et al.* (2013) reported this value as 56.39 kJ/mol for grapes.

Conclusion

The drying behavior of Savory leaves was investigated in a forced conductive dryer at three drying air temperatures, three different air flow velocities and three levels of relative humidity. In order to explain the drying behavior of Savory leaves, nine mathematical models in the literature were employed and fitted to the experimental data. According to the statistical analysis, the Midilli *et al.* model was considered as the best models describing the drying curves of savory leaves by higher the correlation coefficient and lower reduced SSE and RMSE. It can be identified that Midilli *et al.* model could describe the drying behavior of savory leaves during drying process at a temperature range 40-80°C, air relative humidity 20-40% and air flow velocity of 1-2 m/s. The effect of air temperature on drying rate and drying time, for higher values of the moisture content, increase in drying air temperature caused to increase in drying rate and, in follows, decreasing drying time. This can be described by the increasing temperature difference between the drying air and the samples and, in follows,

accelerating moisture migration. The effect of air relative humidity on drying rate and drying time, decreasing the relative humidity increases drying rate change by the drying time. . In other words, at low relative humidity, heat and mass transfer is high and water loss is excessive (higher difference in partial pressure of water between the air and samples). Also the effect of air flow velocity on drying time, with increasing of air velocity, increasing drying rate and, in follows, decreasing drying time were observed. The moisture diffusion coefficient varied between 6.76143×10^{-12} and 1.57018×10^{-10} m²/s for the given temperature range in different air dryer conditions and corresponding activation energy varied between 42.07 and 44.74189 kJ/mol in different air dryer conditions.

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