



processes of various vegetables and agro-based products such as chicory root (Lee *et al.* 2004), basil (Ozcan *et al.* 2005), aromatic plants (Akpınar 2006), chard leaves (Alibas 2006), tea leaves (Ghodake *et al.* 2006), table olive (Demir *et al.* 2007), rehmannia (Rhim *et al.* 2007), fever leaves (Sobukola and Dairo 2007), rosemary leaves (Arslan and Ozcan 2008), onion (Lee and Kim 2008), barberries (Aghbashlo *et al.* 2009) and spinach leaves (Doymaz 2009). However, there is no information about the drying process of thyme in the literature. The main objectives of this research were to investigate the effect of temperature on the air drying kinetics of thyme, to assess the ability of selected drying models to quantify the moisture removal behavior in thyme, and to calculate the effective diffusivity and activation energy.

## MATERIALS AND METHODS

### Experimental Material

Fresh thyme samples (*T. vulgaris* L.) were obtained directly from a grower at Iskenderun, Hatay, Turkey. The 400-g samples were packed in plastic bags and stored in a refrigerator at 4°C to keep the plants as fresh as possible for drying experiments. The average initial moisture content of thyme samples was  $74.18 \pm 0.5\%$  (w.b.), as determined by vacuum drying at 70°C for 24 h (AOAC 1990).

### Experimental Procedure

The drying of thyme was carried out in an experimental drying cabinet, designed and manufactured at the API & PASILAC Limited of Carlisle, Cumbria, U.K. The cabinet dryer is described previously by Doymaz (2005). Experiments were conducted at air temperatures of 40, 50 and 60°C, air velocity of 2.0 m/s, relative humidity of 14–45% and equilibrium moisture content of 0.01705–0.01895 kg water/kg dry matter. The relative humidity and equilibrium moisture content values were determined using wet and dry bulb temperatures obtained from the psychrometric chart. The air velocity was measured directly in the drying chamber with an anemometer (Lutron AM-4201, Taipei, Taiwan) and flowed horizontal to the bed.

The dryer was run without the sample for about 1 h to set the desired conditions before each drying experiment. Then, the thyme samples (simple thickness:  $2.0 \pm 0.1$  cm) were distributed uniformly into the drying tray. The sample weight was kept constant at 60 g ( $\pm 0.5$  g) for all runs. The drying samples were weighed at 15-min intervals during drying by an electronic balance (model BB3000, Mettler-Toledo AG, Greifensee, Switzerland), having an accuracy of  $\pm 0.1$  g. The drying process was continued until the final moisture content of about  $10 \pm 0.5\%$  (w.b.) from an initial value of  $74.18 \pm 0.5\%$  (w.b.). The product was cooled in room tem-

perature for 10 min after drying and kept in airtight glass jars. The sample weight loss during drying was converted into the moisture content and expressed as kg water/kg dry matter.

### Mathematical Modeling of Drying Curves

The moisture ratio (*MR*) of thyme was obtained using the equation below (Lee and Kim 2008):

$$MR = \frac{M_t - M_e}{M_0 - M_e} \quad (1)$$

where  $M_t$  is the moisture content at anytime (kg water/kg dry matter),  $M_0$  is the initial moisture content (kg water/kg dry matter) and  $M_e$  is the equilibrium moisture content of samples (kg water/kg dry matter).

The drying rate (*DR*) of thyme was calculated using Eq. (2) (Lee and Kim 2008):

$$DR = \frac{M_{t+dt} - M_t}{dt} \quad (2)$$

where  $M_{t+dt}$  is the moisture content at  $t + dt$  (kg water/kg dry matter), and  $t$  is the drying time (min).

Drying curves were fitted to 12 thin-layer drying models that are widely used in the scientific literature to describe the kinetics of the drying process. The selected thin-layer drying models are identified in Table 1. The regression analysis was performed using Statistica 6.0 software (Statsoft Inc., Tulsa, OK), which is based on the Levenberg–Marquardt algorithm. The four criteria of statistical analysis have been used to evaluate the adjustment of the experimental data to the different models: the coefficient of determination ( $R^2$ ), mean relative percent error (P), reduced chi-square ( $\chi^2$ ) and RMSE. These parameters can be calculated as

$$P = \frac{100}{N} \sum_{i=1}^N \frac{|MR_{exp,i} - MR_{pre,i}|}{MR_{exp,i}} \quad (3)$$

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

$$RMSE = \left[ \frac{1}{N} \sum_{i=1}^N (MR_{pre,i} - MR_{exp,i})^2 \right]^{1/2} \quad (5)$$

where  $MR_{exp,i}$  and  $MR_{pre,i}$  are the experimental and predicted dimensionless *MR*, respectively,  $N$  is the number of observations, and  $z$  is the number of constants. The best model describing the drying characteristics of samples was chosen as the one with the highest  $R^2$ , the least P,  $\chi^2$  and RMSE (Lee *et al.* 2004; Sacilik *et al.* 2006; Lee and Kim 2008).

**TABLE 1.** MATHEMATICAL MODELS APPLIED TO THE DRYING CURVES

Model number and name	Model	Reference
1. Newton	$MR = \exp(-kt)$	Ozcan <i>et al.</i> (2005); Roberts <i>et al.</i> (2008).
2. Henderson and Pabis	$MR = a \exp(-kt)$	Ghodake <i>et al.</i> (2006)
3. Modified Henderson and Pabis	$MR = a \exp(-kt) + b \exp(-gt) + c \exp(-ht)$	Karathanos (1999).
4. Page	$MR = \exp(-kt^n)$	Sobukola and Dairo (2007); Hassan-Beygi <i>et al.</i> (2009).
5. Logarithmic	$MR = a \exp(-kt) + c$	Xanthopoulos <i>et al.</i> (2007); Wang <i>et al.</i> (2007).
6. Two-term	$MR = a \exp(-k_0t) + b \exp(-k_1t)$	Sacilik <i>et al.</i> (2006).
7. Two-term exponential	$MR = a \exp(-kt) + (1 - a) \exp(-kat)$	Sharaf-Eldeen <i>et al.</i> (1980)
8. Approximation of diffusion	$MR = a \exp(-kt) + (1 - a) \exp(-kbt)$	Yaldiz and Ertekin (2001); Sacilik <i>et al.</i> (2006).
9. Verma <i>et al.</i>	$MR = a \exp(-kt) + (1 - a) \exp(-gt)$	Verma <i>et al.</i> (1985); Akpinar (2006)
10. Wang and Singh	$MR = 1 + at + bt^2$	Demir <i>et al.</i> (2007)
11. Midilli <i>et al.</i>	$MR = a \exp(-kt^n) + bt$	Midilli <i>et al.</i> (2002); Arslan and Ozcan (2008).
12. Parabolic	$MR = a + bt + ct^2$	Sharma and Prasad (2004)

**Calculation of the Effective Moisture Diffusivity**

Drying of most food materials occurs in the falling rate period, and moisture transfer during the drying process is controlled by internal diffusion. Fick’s second diffusion equation (Eq. 6) has been widely used to describe the drying process during the falling rate period for agricultural materials (Roberts *et al.* 2008):

$$\frac{\partial M}{\partial t} = D_{eff} \nabla^2 M \tag{6}$$

The diffusion equation (Eq. 6) is solved for an infinite slab, assuming unidimensional moisture movement volume change, constant temperature and diffusivity coefficients, and negligible external resistance (Crank 1975):

$$MR = \frac{8}{\pi^2} \sum_{n=0}^{\infty} \frac{1}{(2n+1)^2} \exp\left(-\frac{(2n+1)^2 \pi^2 D_{eff} t}{4L^2}\right) \tag{7}$$

where  $D_{eff}$  is the effective diffusivity coefficient ( $m^2/s$ ),  $L$  is the half thickness of the slab (m), and  $n$  is the positive integer. For long drying times, Eq. (7) simplifies to a limiting form of the diffusion equation as follows:

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

The slope ( $K$ ) is calculated by plotting  $\ln(MR)$  versus time according to Eq. (8).

$$K = \frac{\pi^2 D_{eff}}{4L^2} \tag{9}$$

**Calculation of Activation Energy**

Effective diffusivity can be related to temperature by Arrhenius expression (Simal *et al.* 2006) such as:

$$D_{eff} = D_0 \exp\left(-\frac{E_a}{R(T+273.15)}\right) \tag{10}$$

where  $D_0$  is the constant in the Arrhenius equation ( $m^2/s$ ),  $E_a$  is the activation energy (kJ/mol),  $T$  is the temperature of air (C), and  $R$  is the universal gas constant (kJ/mol K). Eq. (10) can be rearranged into the form of:

$$\ln(D_{eff}) = \ln(D_0) - \frac{E_a}{R(T+273.15)} \tag{11}$$

**Rehydration Test**

Rehydration was carried out by immersing dried water thyme in distilled water maintained at three different temperatures, namely, 20, 40 and 60C ( $\pm 1C$ ). About 2 g of the dried product was added to 400 mL of distilled water in a 400-mL beaker. Samples were removed after 4 h, dried with tissue paper and weighed using an electronic digital balance (Precisa, model XB220A, Precisa Instruments AG, Dietikon, Switzerland). The rehydration ratio was calculated as follows:

$$\text{Rehydration ratio} = \frac{\text{Weight of rehydrated thyme samples (g)}}{\text{Weight of dried thyme samples (g)}} \tag{12}$$

**RESULTS AND DISCUSSION**

**Effect of Air Drying Temperature**

The typical drying curves for thyme at air temperatures of 40, 50 and 60C are shown in Fig. 1 for an air velocity of 2.0 m/s. As expected for a thermally activated process, as temperature was increased, the initial slope of the drying curve increased, and the drying time taken to achieve final moisture content consequently decreased (Fig. 1). The drying time required to reduce the moisture content to any given level was dependent on the

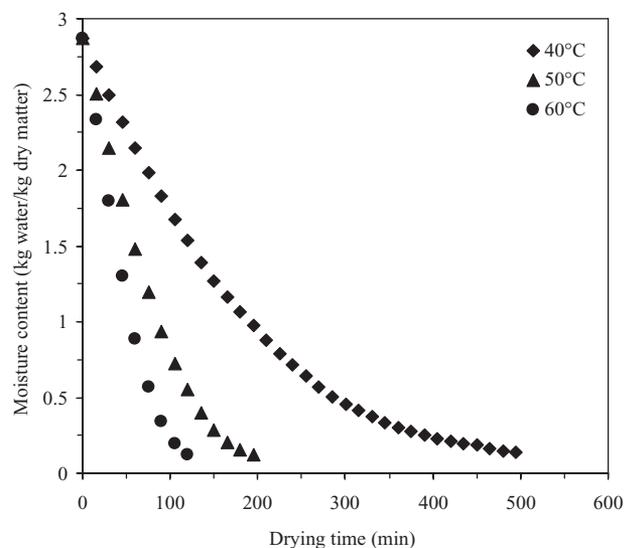


FIG. 1. VARIATIONS OF MOISTURE CONTENT AS A FUNCTION OF DRYING TIME FOR DIFFERENT AIR TEMPERATURES

drying condition, being the highest at 40C and the lowest at 60C. The drying times of thyme were 495, 195 and 120 min, respectively, in relation to the temperature levels of 40, 50 and 60C applied in drying practices. The drying time at 60C was shortened by 1.5 and 4.12 times compared with the drying process realized at 50C and 40C, respectively. Consequently, the effect of air temperature has been reflected in drying time. Similar results were reported by Khazaei *et al.* (2008), Alibas (2006), Rhim *et al.* (2007) and Lee and Kim (2008).

The DRs (kg water/(kg dry matter × min)) obtained in unit time under different temperatures are given in Fig. 2. DR increased with the increase of air drying temperature, and the highest values of DR were obtained during the experiment at 60C. From the curves, it is also clearly shown that the drying temperature had a significant effect on the DR, and no con-

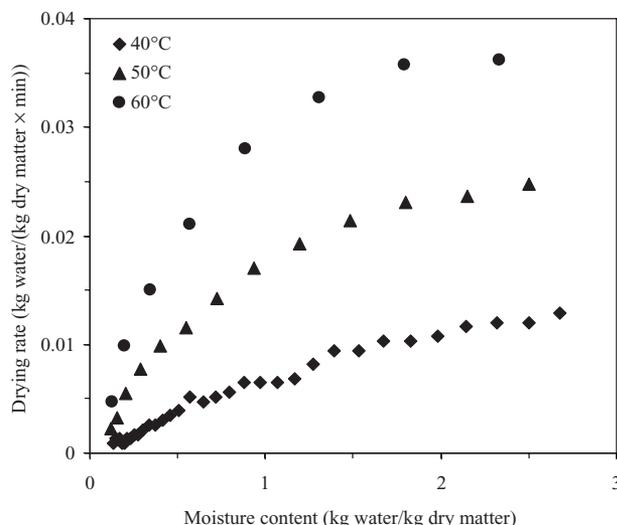


FIG. 2. VARIATION OF DRYING RATE AS A FUNCTION OF MOISTURE CONTENT FOR DIFFERENT AIR TEMPERATURES

stant rate drying period was observed. All the drying processes occurred in the falling rate drying period. This indicates that diffusion is the most likely physical mechanism governing moisture movement in the thyme particles. The results were generally in agreement with some literature studies on drying of various food products (Panchariya *et al.* 2002; Simal *et al.* 2006; Rhim *et al.* 2007; Lee and Kim 2008).

### Fitting of the Drying Curves

For the drying conditions of each run, the experimental data fitted to all models enlisted in Table 1 and tested for  $R^2$ , P,  $\chi^2$  and RMSE values. The statistical analysis values are summarized in Tables 2–4. All the models gave consistently high coefficient of determination ( $R^2$ ) values in the range of 0.9748–0.9998. This indicates that all the models could satis-

TABLE 2. CURVE FITTING CRITERIA FOR THE MATHEMATICAL MODELS AND PARAMETERS AT 40C OF DRYING AIR

Model number	Model constants	$R^2$	P	$\chi^2$	RMSE
1	k: 0.0057	0.9934	14.3724	0.00055	0.12246
2	a: 1.0482, k: 0.0060	0.9962	10.3605	0.00033	0.09043
3	a: -13.3284, k: 0.0037, b: 7.1763, g: 0.0038, c: 7.1759, h: 0.0038	0.9989	6.0419	0.00010	0.04541
4	k: 0.0025, n: 1.1508	0.9997	2.3626	0.00001	0.02082
5	a: 1.0823, k: 0.0052, c: -0.0548	0.9986	6.9295	0.00012	0.05335
6	a: -5.4067, $k_0$ : 0.0096, b: 6.4038, $k_1$ : 0.0087	0.9998	2.4469	0.00001	0.01674
7	a: 0.0019, k: 3.0363	0.9932	14.6151	0.00059	0.12448
8	a: -0.1812, k: 0.0248, b: 0.2656	0.9993	4.2331	0.00005	0.03629
9	a: 3.3155, k: 0.0037, g: 0.0031	0.9983	7.4772	0.00014	0.05711
10	a: -0.0043, b: 0.0001	0.9977	10.2006	0.00019	0.06477
11	a: 0.9903, k: 0.0023, n: 1.1678, b: 0.0001	0.9998	2.2773	0.00001	0.02051
12	a: 0.9827, b: -0.0042, c: 0.0001	0.9981	8.8171	0.00016	0.05911

**TABLE 3.** CURVE FITTING CRITERIA FOR THE MATHEMATICAL MODELS AND PARAMETERS AT 50C OF DRYING AIR

Model number	Model constants	R <sup>2</sup>	P	χ <sup>2</sup>	RMSE
1	k: 0.0129	0.9776	34.4017	0.00240	0.16139
2	a: 1.0712, k: 0.0138	0.9842	28.4083	0.00183	0.13927
3	a: -2.8339, k: 0.0292, b: 3.7841, g: 0.0222, c: 0.0497, h: 0.9669	0.9991	4.7679	0.00014	0.02822
4	k: 0.0029, n: 1.3303	0.9993	3.6278	0.00008	0.02527
5	a: 1.2067, k: 0.0098, c: -0.1714	0.9957	13.2205	0.00054	0.06713
6	a: -23.7567, k <sub>0</sub> : 0.0248, b: 24.7517, k <sub>1</sub> : 0.0240	0.9990	5.7464	0.00013	0.03089
7	a: 0.0019, k: 6.5737	0.9772	34.7292	0.00265	0.16259
8	a: -7.3976, k: 0.0256, b: 0.9043	0.9990	5.8987	0.00012	0.03036
9	a: 3.8422, k: 0.0063, g: 0.0047	0.9950	13.9284	0.00063	0.06821
10	a: -0.0097, b: 0.0001	0.9984	3.7109	0.00018	0.03183
11	a: 0.9957, k: 0.0031, n: 1.3096, b: -0.0001	0.9994	3.5900	0.00007	0.02201
12	a: 1.0201, b: -0.0101, c: 0.0001	0.9990	3.6111	0.00012	0.02803

factorily describe the air drying of thyme. Among the thin-layer models, the Midilli *et al.* model obtained the highest R<sup>2</sup> values and the lowest P, χ<sup>2</sup> and RMSE values in the temperature range of the study. It is clear that the R<sup>2</sup>, P, χ<sup>2</sup> values of this model were changed between 0.9994 and 0.9998, 2.2773 and 6.1428, 0.00001 and 0.00008, and 0.01785 and 0.02201, respectively. Thus, this model may be assumed to present the thin-layer drying behavior of the thyme. Figure 3 shows the variation of experimental and predicted moisture ratios for thyme samples dried at 40, 50 and 60C. It can be seen that the proposed model provided a good conformity between experimental and predicted moisture ratios. Similar findings were reported by McMinn (2006) for lactose powder drying and Hacıhafızoglu *et al.* (2008) for rough rice drying.

**Determination of Effective Moisture Diffusivity**

The values of effective moisture diffusivity for all the drying conditions, calculated from Eq. (9), are presented in Table 5, as well as the obtaining values between 1.097 and

5.991 × 10<sup>-9</sup> m<sup>2</sup>/s. As expected, the effective moisture diffusivity values increased greatly with the increase in drying temperature because of increase in the vapor’s pressure inside the samples. The values of D<sub>eff</sub> obtained from this study lie within the general range of 10<sup>-12</sup>–10<sup>-8</sup> m<sup>2</sup>/s for drying of food materials (Zogzas *et al.* 1996). Similar results are found to correspond well with those existing in the literature, such as 0.659–1.927 × 10<sup>-9</sup> m<sup>2</sup>/s for spinach leaves (Doymaz 2009), 5.30–17.73 × 10<sup>-10</sup> m<sup>2</sup>/s for aloe vera (Vega *et al.* 2007), 1.345–2.658 × 10<sup>-8</sup> m<sup>2</sup>/s for onion slices (Lee and Kim 2008), 1.021–10.44 × 10<sup>-9</sup> m<sup>2</sup>/s for Mexican tea leaves (Ethmane Kane *et al.* 2008) and 1.744–4.992 × 10<sup>-9</sup> m<sup>2</sup>/s for nettle and mint leaves (Kaya and Aydin 2009). These values are consistent with the present estimated D<sub>eff</sub> values for thyme.

**Determination of Activation Energy**

The natural logarithm of D<sub>eff</sub> as a function of the reciprocal of absolute temperature (Eq. 11) was plotted in Fig. 4. The result shows a linear relationship derived from the Arrhenius-type

**TABLE 4.** CURVE FITTING CRITERIA FOR THE MATHEMATICAL MODELS AND PARAMETERS AT 60C OF DRYING AIR

Model number	Model constants	R <sup>2</sup>	P	χ <sup>2</sup>	RMSE
1	k: 0.0020	0.9751	40.9145	0.00301	0.13411
2	a: 1.0576, k: 0.0211	0.9800	36.2479	0.00276	0.12838
3	a: -11.4584, k: 0.0400, b: 6.2264, g: 0.0373, c: 6.2268, h: 0.0373	0.9985	9.9742	0.00046	0.030772
4	k: 0.0044, n: 1.3725	0.9990	6.2775	0.00012	0.02144
5	a: 1.2627, k: 0.0136, c: -0.2389	0.9958	15.9065	0.00067	0.05931
6	a: -25.4717, k <sub>0</sub> : 0.0393, b: 26.4666, k <sub>1</sub> : 0.0380	0.9958	9.9448	0.00027	0.03071
7	a: 0.0018, k: 10.8034	0.9748	41.6364	0.00349	0.13554
8	a: -8.9491, k: 0.0403, b: 0.9154	0.9985	10.0630	0.00023	0.02997
9	a: -2.5443, k: 0.0063, g: 0.0090	0.9956	15.8767	0.00070	0.05552
10	a: -0.0014, b: 0.0001	0.9984	6.9783	0.00021	0.03125
11	a: 0.9963, k: 0.0052, n: 1.3205, b: -0.0002	0.9995	6.1428	0.00008	0.01785
12	a: 1.017, b: -0.0153, c: 0.0001	0.9989	5.3508	0.00017	0.02820

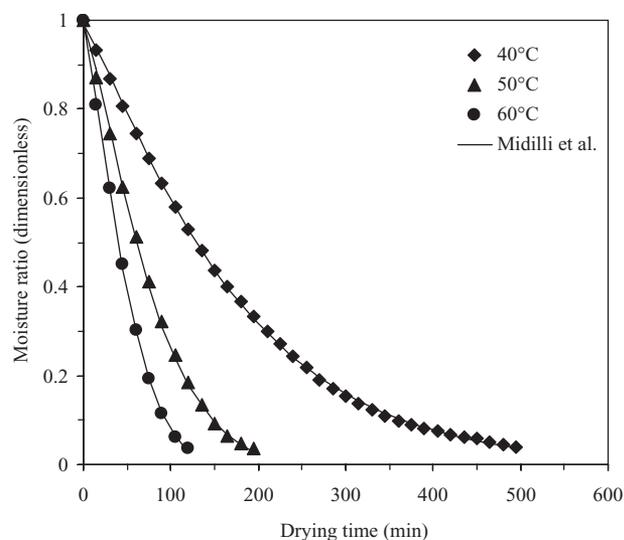


FIG. 3. EXPERIMENTAL AND SIMULATED CURVES USING THE MIDILLI ET AL. MODEL FOR DIFFERENT AIR TEMPERATURES

equation. From the slope of the straight line described by the Arrhenius equation, the activation energy was found to be 73.84 kJ/mol. The comparison with literature values for various vegetables is shown in Table 6. It is higher than the activation energies of okra drying (Doymaz 2005), rehmannia drying (Rhim *et al.* 2007), avishan drying (Khazaei *et al.* 2008) and onion slices drying (Lee and Kim 2008), and lower than the activation energies of mint leaves drying (Park *et al.* 2002), black tea drying (Panchariya *et al.* 2002) and fever leaves drying (Sobukola and Dairo 2007).

TABLE 5. VALUES OF EFFECTIVE MOISTURE DIFFUSIVITY OBTAINED FOR THYME AT DIFFERENT TEMPERATURES

Temperature (C)	Effective diffusivity ( $D_{eff}$ ), $m^2/s$
40	$1.097 \times 10^{-9}$
50	$3.342 \times 10^{-9}$
60	$5.991 \times 10^{-9}$

TABLE 6. ACTIVATION ENERGY FOR DRYING OF FOOD MATERIALS

Material	$E_a$ (kJ/mol)	References
Black tea	406.02	Panchariya <i>et al.</i> (2002)
Mint	82.93	Park <i>et al.</i> (2002)
Fever leaves	80.78	Sobukola and Dairo (2007)
Thyme	73.84	Present work
Okra	51.26	Doymaz (2005)
Rehmannia	47.14	Rhim <i>et al.</i> (2007)
Avishan	46.86	Khazaei <i>et al.</i> (2008)
Onion	31.36	Lee and Kim (2008)

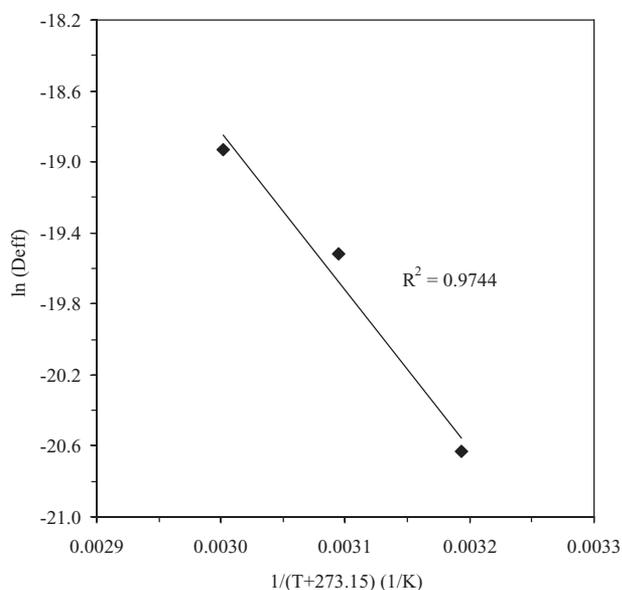


FIG. 4. INFLUENCE OF AIR TEMPERATURE ON THE EFFECTIVE MOISTURE DIFFUSIVITY

### Effect of Temperature on Rehydration Ratio

Rehydration ratio is widely used as a parameter for dried sample quality. They indicate the physical and chemical changes during drying as influenced by processing conditions, sample pretreatment and composition (Feng and Tang 1998). Rehydration of thyme samples was performed at different temperatures (20, 40 and 60C). The values obtained, calculated from Eq. (12), are shown in Fig. 5. The rehydration

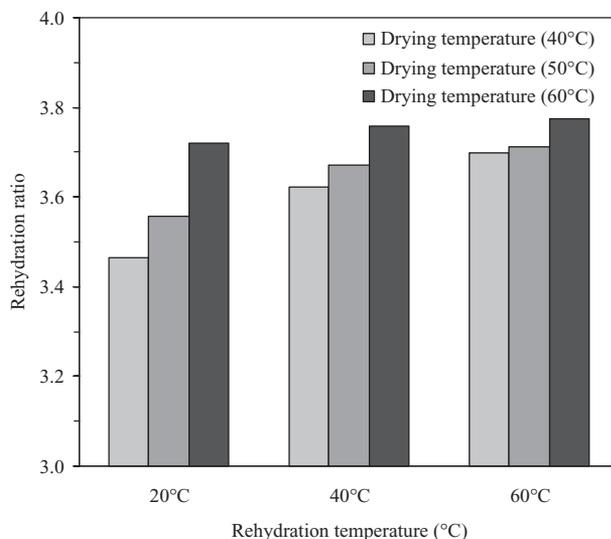


FIG. 5. REHYDRATION RATIO VERSUS DIFFERENT REHYDRATION TEMPERATURES

ratio was affected significantly by the drying temperatures at all the selected rehydration temperatures. The rehydration ratio increased as the drying temperature increased. Rehydration ratio at 60C was observed more rapidly than at 20 and 40C (3.722 kg water/kg dry matter for dried sample at 60C). Rehydration at high temperatures improves because of the effect of temperature on cell wall and tissue (Singh *et al.* 2008).

## CONCLUSIONS

The results showed that the drying characteristics of thyme were greatly affected by air temperature. The increase in air temperature significantly reduced the drying time of thyme. The rehydration ratio increased as the drying temperature was increased. In drying, curves of samples did not show a constant rate drying period under the experimental process employed and showed only a falling rate period. Twelve widely used thin-layer drying models were fitted to data obtained from drying experiments. The Midilli *et al.* model showed a good fit curves than the other models. The values of effective diffusivity for drying at 40–60C of air temperature and at 2.0 m/s of air velocity ranged from 1.097 to  $5.991 \times 10^{-9}$  m<sup>2</sup>/s. The effective diffusivity increased with the increasing air temperature. Temperature dependence of the diffusivity coefficients was described by Arrhenius-type relationship. The activation energy for moisture diffusion was found as 73.84 kJ/mol.

## NOMENCLATURE

a, b, c	drying coefficients
$D_{eff}$	effective moisture diffusivity (m <sup>2</sup> /s)
g, h, k, k <sub>0</sub> , k <sub>1</sub>	drying constants
K	slope
$MR_{exp,i}$	experimental moisture ratio
$MR_{pre,i}$	predicted moisture ratio
$M_e$	equilibrium moisture content (kg water/kg dry matter)
$M_0$	initial moisture content (kg water/kg dry matter)
$M_t$	moisture content at anytime (kg water/kg dry matter)
$M_{t+dt}$	moisture content at t + dt (kg water/kg dry matter)
n	number of observations
N	constant, positive integer
P	mean relative percent error
$R^2$	determination of coefficient
RMSE	root mean square error
t	drying time (min)
z	number of coefficients and constants
$\chi^2$	reduced chi-square

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