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Research Article

The effect of vacuum on the drying kinetics and mathematical modelling of blueberries

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ABSTRACT

Blueberry is a small fruit, which has a very high amount of antioxidant substances. It is consumed either in fresh or dried form. In this study, the effect of vacuum on the oven thin-layer drying of blueberries was investigated. The kinetic parameters of effective moisture diffusivity (D_{eff}) and activation energy (E_a) values were calculated. Moreover, the drying curves were modelled with the most known mathematical modelling equations given in the literature. For oven and vacuum oven drying, the drying temperatures were selected as 60, 70 and 80°C. The drying times were seen to decrease by increasing the drying temperature and with the effect of vacuum. The highest drying time was observed as 420 minutes in oven drying at 60°C. The effect of vacuum decreased this drying time to 240 minutes. D_{eff} values were calculated to elucidate the underlying mass transfer mechanisms by using Fick's 2nd law, which were found to be between 1.14-2.83×10^{-9} m^2/s for oven drying and increased to 2.54-4.33×10^{-9} m^2/s by the addition of vacuum. Likewise, activation energy was increased from 25.93 to 43.98 kJ/mol by vacuum addition. Twelve mathematical models were applied to the drying curve data and among them, Alibas model for both oven drying and vacuum-assisted oven drying gave the coefficient of determination (R^2) values that were higher than 0.997. As a result, the vacuum addition was seen to yield lower drying times and higher kinetic parameters for the oven drying of blueberries.

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INTRODUCTION

Drying process occupies a substantial place in food industry. The main objective of drying process in food industry is to preserve foods and to increase their shelf lives, by decreasing their moisture contents in order to inhibit the activities of microorganisms. The demand for dried foods is particularly increasing due to their practical use, especially considering the prominently reduced weight and volume through the drying process. Moreover, it is not necessary to use refrigeration systems for transport and store dried foods. Consequently, it also reduces the transport and storage costs. Among the traditional methods employed for drying, oven drying method offers the most simple and easy application, being even suitable for

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domestic implementations. Oven drying provides a faster, more hygienic and homogeneous drying than sun or solar drying, methods of which can also be stated among the traditional drying methods. Furthermore, the costs of installation, maintenance and repair are lower for oven drying [1-5]. Sometimes, traditional drying methods are assisted with vacuum addition. Introducing such a vacuum assistance protects the food against oxidation, hence the nutritional values, taste, texture and color of the product are preserved [6].

Blueberries (*Vaccanium corymbosum*) are small, perishable fruits with blue color. Blueberries are cultivated mostly in North and South America. They have highly economic value, including many healthy nutrients and polyphenols. But they are seasonal fruits, having a very short shelf life. In nutritional values point of view, blueberries have abundant amount of water, glucose and fructose sugars. Blueberries also contain high amount of organic acids of citric and ascorbic acid, phosphorus, potassium and magnesium minerals and fibers. They are known as very good antioxidant sources and thus have the potential to reduce the adverse impacts of some diseases like memory loss, heart disease and cancer [7,8]. Research has shown that there are fifteen different anthocyanins in blueberries such as monoarabinosides, monoglucosides and monogalactosides of cyanidin, peonidin, delphinidin, petunidin and malvidin [9]. The polyphenol content of blueberries is one of the highest amount among fruits and vegetables [10]. By 2020, the top five blueberry producers were USA (294000 tons) followed by Peru (180300 tons), Canada (146370 tons), Poland (55300 tons) and Mexico (50293 tons) [11].

In the literature, although there are numerous articles focusing on the antioxidant capacities and nutritional contents of blueberries, studies investigating their drying process is still scarce. Among these studies present, Shi et al. worked on the infrared drying kinetics of blueberries [8]; Vega-Gálvez et al. worked on the drying characteristics of blueberries by using hot-air method [12]; Zielinska et al. investigated the drying of blueberries in hot-air and microwave combined system [13] and MacGregor studied wild blueberry drying in a packed bed dryer, in which the effects of air velocity, air temperature and berry diameter on drying were investigated [14]. As it is seen from the literature, a detailed research on the drying kinetics of blueberries and the conductance of its mathematical modeling is worth investigation, since the majority of the published articles involve examining their antioxidant contents. In addition, the effect of vacuum assistance for oven drying is another subject that has not been sufficiently investigated up to date. Furthermore, the application of the drying curve data to a wide range of mathematical models has not been fully carried out in the literature. From this aspect, in this study, the investigation of the drying kinetic parameters of effective moisture diffusivities (D_{eff}) and activation energy values (E_a) for oven drying of blueberries, with and without the assistance of vacuum was studied. It was also aimed to determine the effect of vacuum on the aforementioned kinetic parameters and drying times. Moreover, the drying curve data was applied to twelve most known mathematical models.

MATERIALS AND METHODS

Samples

Chile origin blueberries were retrieved from a local market in Istanbul, Turkey on November 2021. Similar sized blueberries were selected (~ 1 cm radius) and cut into two parts horizontally before the experiments, to investigate the thin-layer diffusion. In each experiment set, 4.40 ± 0.30 g of blueberries were dried. Prior to drying, by using AOAC method, the initial moisture content (M_0) of the blueberries was determined [15]. For the M_0 determination, conventional hot air-drying oven (KH-45, Kenton, Guangzhou, China) was used at 105°C for 2 hours. Accordingly, the initial moisture content of the blueberries was determined on a wet basis as 84.33%, which was 5.38 kg water/kg dry matter.

Experimental Method

In the first stage of the dying experiments, oven (EV-018, Nüve, Ankara, Turkey) drying, at drying temperatures of 60, 70, and 80°C were tested. At every 15 minutes, samples were weighed to determine their moisture contents. Sample weights were measured by using a digital balance (AS 220.R2, Radwag, Radom, Poland), which had a weighting accuracy of 0.001 g. In the second stage of the experiments, vacuum assistance was supplied on the same oven of Nüve EV-018, by using a vacuum pump KNF N022AN.18 (KNF, Freiburg, Germany) which operated at 100 W and 40 kHz frequency. The pressure inside the oven was measured as 0.3 atm during drying. In 15 minute intervals, the samples were weighed after the pressure valve connected to the oven was opened and the pressure inside the oven came into equilibrium with room pressure. Three parallel drying experiments were conducted in each drying method.

Drying Curves

In order to construct the drying curves of the blueberries, the moisture contents (M), drying rates (DR) and moisture ratios (MR) must be determined. These parameters were calculated by using Eq. (1), (2) and (3) [16-18] given below:

$$M = \frac{m_w}{m_d} \quad (1)$$

In Eq. (1) M represents the content of moisture in kg water/kg dry matter, m_w represents the water content of the blueberries in kg, and m_d is the dry matter content in kg.

$$DR = \frac{M_{t+dt} - M_t}{t_{i+1} - t_i} \quad (2)$$

In Eq. (2) DR represents the drying rate in kg water/kg dry matter × min, t is the drying time in minutes and M_{t+dt} is the amount of moisture during the time $t+dt$ in kg water/kg dry matter.

$$MR = \frac{M_t - M_e}{M_o - M_e} \quad (3)$$

In the above equation MR represents the moisture content (dimensionless), M_t, M_i, and M_e represent the amount of moisture at any time, at initial conditions and at balance, respectively. Since the moisture levels in the balance are very low compared to the initial and at any moment humidity values, M_e is neglected in the calculations [19].

Effective Moisture Diffusivity and Activation Energy Calculations

Drying of food materials is a function of internal diffusion and usually occurs in the falling rate period. To describe moisture diffusion, generally Fick's 2nd law of diffusion is used. While solving this equation, several assumptions were made. The shrinkage was neglected, the diffusion coefficient was accepted as constant, and it was assumed that the mass transfer occurred symmetrically with respect to the center only by diffusion. The applicable equation of Fick's 2nd law for the unsteady state condition in a 2L thick thin layer is given in Eq. (4) [20,21]:

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

In Eq. (4) n is the positive integer, t is the time in seconds, D_{eff} in m²/s, and L is the half-thickness of the sample in meters. For long drying times, n is assumed as 1 [20,21]. Thus Eq. (4) can be simplified to Eq. (5):

$$\ln(MR) = \ln\left(\frac{8}{\pi^2}\right) - \left(\pi^2\frac{D_{eff}\times t}{4L^2}\right) \quad (5)$$

By using the equation given above, D_{eff} can be calculated from the slope of the ln(MR) versus t plot. On the other hand, the relation between the D_{eff} and temperature variation can be expressed by the Arrhenius equation, which is shown in Eq. (6) [22]:

$$D_{eff} = D_o\exp\left(-\frac{E_a}{R(T+273.15)}\right) \quad (6)$$

In the aforementioned equation D_0 is the pre-exponential factor in m²/s, E_a in kJ/mol, R is the universal gas constant in kJ/mol × K, and T is the drying temperature in °C. E_a can be calculated from the slope of the plot of ln(D_{eff}) versus 1/T.

Mathematical Modelling

The mathematical model equations that were used in this study are presented in Table 1. In the modelling process, Statistica software (Statsoft Inc., Tulsa, OK) was used for the nonlinear Levenberg-Marquardt procedure regressions. The coefficient of determination (R^2), reduced chi-square (χ^2) and root mean square error (RMSE) values were obtained from Eq. (7) through Eq. (9) [23-28]:

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

$$\chi^2 = \frac{\sum_{i=1}^{N}(MR_{exp,i} - MR_{pre,i})^2}{N-n} \quad (8)$$

$$\text{RMSE} = \sqrt{\frac{\sum_{i=1}^{N}(MR_{exp,i} - MR_{pre,i})^2}{N}} \quad (9)$$

The best model was selected as the one giving the highest R^2, along with the lowest χ^2 and RMSE values [19,29].

Table 1. Mathematical models and equations

Model Equation	Model Name
$MR = \exp(-k_1\times t/(1+k_2\times t))$	Aghbaslo et al.
$MR = a\times\exp((-k\times t^n)+b\times t)+g$	Alibas
$MR = a\times\exp(-k\times t)$	Henderson & Pabis
$MR = a\times\exp(-k\times t + b\sqrt{t}) + c$	Jena et al.
$MR = \exp(-k\times t)$	Lewis
$MR = a\times\exp(-k\times t) + c$	Logarithmic
$MR = a\times\exp(-k\times t^n) + b\times t$	Midilli & Kucuk
$MR = \exp(-k\times t^n)$	Page
$MR = a+b\times t+c\times t^2$	Parabolic
$MR = a\times\exp(-kt)+(1-a)\times\exp(-k\times a\times t)$	Two-Term Exponential
$MR = a\times\exp(-kt)+(1-a)\times\exp(-g\times t)$	Verma et al.
$MR = 1+a\times t + b\times t^2$	Wang & Singh

*a, b, c, g are coefficients and n is the drying exponent specific to each equation; k, k1, k2 are the drying coefficients specific to each equation; t is the time.

RESULTS AND DISCUSSION

Drying Curves

The drying curves and the drying rate curves of oven and vacuum assisted oven drying are given in Figure 1 and Figure 2, respectively. As seen in Figure 1, from M_0 of 5.3800 kg water/kg dry matter, M_f of the oven drying were decreased to 0.3102, 0.2514 and 0.1328 for the drying temperatures of 60, 70 and 80°C, respectively. The drying times were obtained as 420, 390 and 210 minutes for the same temperatures. On the other hand, for the vacuum assisted oven drying, M_f were decreased to 0.1468, 0.1381 and 0.1167 for the drying temperatures of 60, 70 and 80°C,

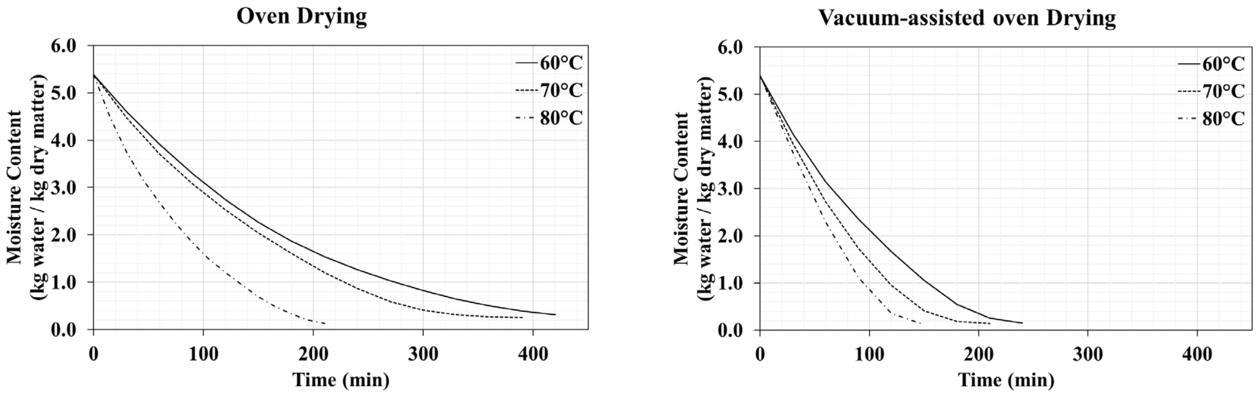

Figure 1. Drying curves of oven and vacuum-assisted oven drying.

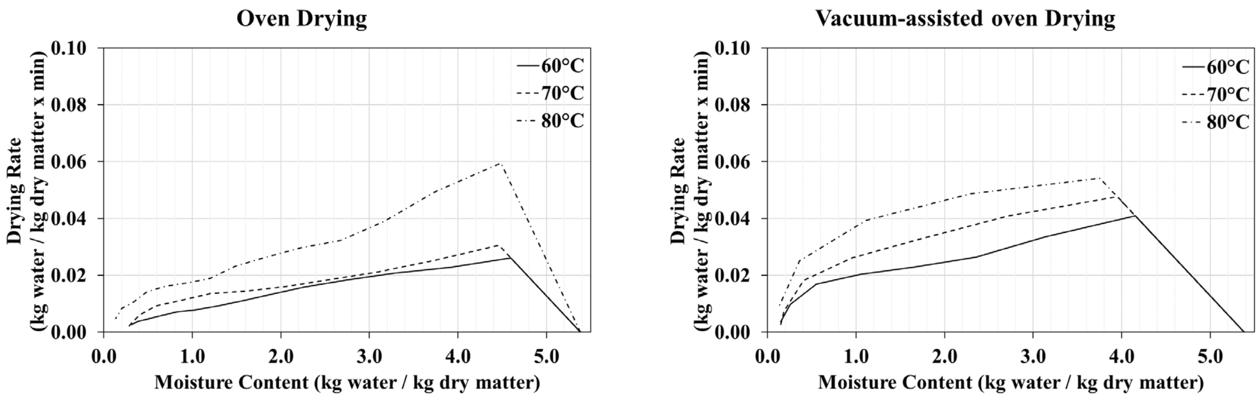

Figure 2. Drying rate curves of oven and vacuum-assisted oven drying.

respectively. With vacuum assistance, the drying times were seen to be decreased further to 240, 210 and 150 minutes at these temperatures. Hence, vacuum's decreasing effect on the M_f and the drying times was clearly observed. Considering this remarkable decrease in the drying times, it can be said that vacuum assisted oven drying offers economic benefits, when compared to solely oven drying of blueberries. These reduced drying times also decrease the amount of energy used for drying, hence offering environmental benefits as well. In addition, as the drying temperatures increased, the drying times were seen to decrease. Similar results were obtained in literature studies [25,29]. From Figure 2, the rising-rate periods and falling-rate periods were observed for both of the drying methods. In the oven drying, the rising rate periods were obtained from the M_0 of 5.3800 kg water/kg dry matter to 4.5965, 4.4650 and 4.4870 kg water/kg dry matter for the drying temperatures of 60, 70 and 80°C, respectively. Then the falling-rate periods were encountered until the M_f. For the vacuum-assisted oven drying, the rising rate periods were obtained from the M_0 of 5.3800 kg water/kg dry matter to 4.1522, 3.9505 and 3.7537 kg water/kg dry matter for the drying temperatures of 60, 70 and 80°C, respectively.

Effective Moisture Diffusivity and Activation Energy Results

D_{eff} values were obtained from Eq. (5) for each method and each drying temperature, from the slope of the ln(MR) versus drying time plots. Obtained equations are presented in Eq. (10) through (15):

$$\text{Oven Drying (60°C)} \rightarrow \ln(MR) = -0.000113t + 0.111101 \quad (R^2 = 0.994692) \tag{10}$$

$$\text{Oven Drying (70°C)} \rightarrow \ln(MR) = -0.000143t + 0.170545 \quad (R^2 = 0.986327) \tag{11}$$

$$\text{Oven Drying (80°C)} \rightarrow \ln(MR) = -0.000279t + 0.258553 \quad (R^2 = 0.963838) \tag{12}$$

$$\text{Vacuum-assisted Oven Drying (60°C)} \rightarrow \ln(MR) = -0.000251t + 0.320189 \quad (R^2 = 0.960166) \tag{13}$$

$$\text{Vacuum-assisted Oven Drying (70°C)} \rightarrow \ln(MR) = -0.000313t + 0.285545 \; (R^2 = 0.976571) \quad (14)$$

$$\text{Vacuum-assisted Oven Drying (80°C)} \rightarrow \ln(MR) = -0.000427t + 0.368105 \; (R^2 = 0.954425) \quad (15)$$

In the oven-drying method D_{eff} values were calculated as 1.14×10^{-9}, 1.45×10^{-9} and 2.83×10^{-9} m^2/s, for the drying temperatures of 60, 70 and 80°C, respectively. On the other hand, by the addition of vacuum, D_{eff} values were increased to 2.54×10^{-9}, 3.17×10^{-9} and 4.33×10^{-9} m^2/s, for the drying temperatures of 60, 70 and 80°C, respectively.

For the calculation of the E_a values, Eq. (6) was used. The plot of $\ln(D_{eff})$ versus $1/T$ was employed for both drying methods, and the obtained equations are given in Eq. (16) and Eq. (17):

$$\text{Oven Drying} \rightarrow \ln(D_{eff}) = -5290T^{-1} - 4.78 \; (R^2 = 0.920) \quad (16)$$

$$\text{Vacuum-assisted Oven Drying} \rightarrow \ln(D_{eff}) = -3119.6T^{-1} - 10.44 \; (R^2 = 0.987) \quad (17)$$

By using these two equatons E_a values were calcuated as 25.93 and 43.98 kJ/mol, for the methods of oven and vacuum-assisted oven drying, respectively. Since the addition of vacuum increased the D_{eff} values, E_a was also observed to increase, as verified by other studies in literature [25,29].

Mathematical Modelling Results

The mathematical modelling results obtained for oven and vacuum assisted oven drying are presented in Table 2. Model equations that resulted with R^2 values less than 0.996 were not included in this table. The maximum R^2 value is obtained in Alibas model at 60°C (0.999968) and the minimum R^2 value (0.997672) is obtained in logarithmic model at 70°C, for oven drying method. For vacuum-assisted oven drying, on the other hand, the maximum R^2 value is obtained in Alibas model at 80°C (0.999784) and the minimum R^2 value (0.996137) is obtained in two-term exponential model at 80°C. Though many of the used models adequately described the data obtained, for oven drying Alibas is the most suitable model with highest R^2 values of 0.999968, 0.999506, 0.999901; lowest χ^2 values of 0.000004, 0.000071, 0.000013; and lowest RMSE values of 0.001658, 0.006734, 0.002944 for 60, 70 and 80°C drying temperatures, respectively. This is followed by the models of Midilli et al. (R^2: 0.999960-0.998238; χ^2: 0.000227-0.000005; RMSE: 0.012722-0.004851), two-term exponential (R^2: 0.999866-0.998121; χ^2: 0.000242-0.000015; RMSE: 0.013139-0.003360), Aghbashlo et al. (R^2: 0.999944-0.998573; χ^2: 0.000144-0.000005; RMSE: 0.011172-0.00217) and logarithmic (R^2: 0.999749-0.997672; χ^2: 0.000272-0.000026; RMSE: 0.014622-0.004603).

On the other hand, for vacuum-assisted oven drying, again Alibas is the most suitable model, which offered the highest R^2 values of 0.999448, 0.999811, 0.999784; the lowest χ^2 values of 0.000129, 0.000057, 0.000160 and the lowest RMSE values of 0.007568, 0.004616, 0.005166 for 60, 70 and 80°C, respectively. This is followed by the models of Aghbashlo et al. (R^2: 0.999784-0.999243; χ^2: 0.000101-0.000040; RMSE: 0.008861-0.005167), Midilli et al. (R^2: 0.999149-0.998726; χ^2: 0.000315-0.000223; RMSE: 0.012004-0.010253), logarithmic (R^2: 0.998634-0.995714; χ^2: 0.001058-0.000213; RMSE: 0.023003-0.011905) and two-term exponential (R^2: 0.996763-0.996137; χ^2: 0.001431-0.000604; RMSE: 0.021839-0.018325). Figure 3 presents the comparison of the calculated MR values and the experimental MR values obtained for the most suitable model of Alibas for both methods. As all of the data lied on the 45° line, it can be concluded that the models fitted could be excellently used to represent the experimental data.

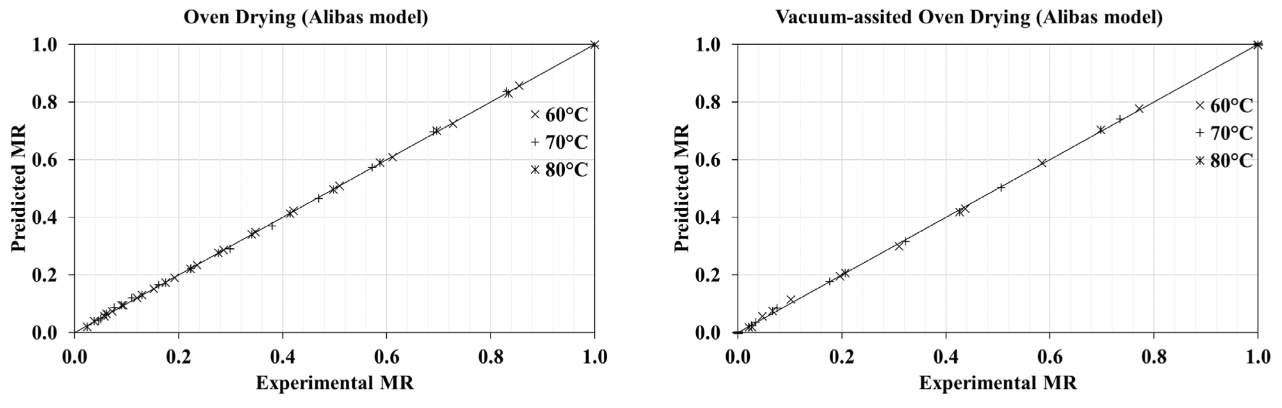

Figure 3. Experimental and calculated *MR* values obtained from the models of Alibas for oven drying and vacuum-assisted oven drying.

Table 2. The obtained mathematical model coefficients and statistical data.

Model	Parameter	Oven Drying			Vacuum-assisted Oven Drying		
		Temperature (°C)			Temperature (°C)		
		60	70	80	60	70	80
Aghbashlo et al.	k_1	0.005225	0.005635	0.010606	0.007577	0.009448	0.010517
	k_2	-0.000565	-0.001005	-0.001523	-0.002106	-0.002833	-0.004379
	R^2	0.999944	0.999015	0.998573	0.999243	0.999507	0.999784
	χ^2	0.000005	0.000106	0.000144	0.000101	0.000074	0.000040
	RMSE	0.002170	0.009514	0.011172	0.008861	0.007470	0.005167
Alibas	a	1.130697	4.91960	3.39385	5.70917	4.53334	5.73171
	k	0.003939	0.00204	0.00604	0.00261	0.00328	0.00325
	n	1.049124	0.99839	0.90821	1.00175	1.06071	1.09687
	b	0.000155	0.00444	0.00406	0.00706	0.00863	0.01437
	g	-0.131191	-3.92463	-2.39333	-4.71150	-3.53495	-4.73280
	R2	**0.999968**	**0.999506**	**0.999901**	**0.999448**	**0.999811**	**0.999784**
	χ2	**0.000004**	**0.000071**	**0.000013**	**0.000129**	**0.000057**	**0.000160**
	RMSE	**0.001658**	**0.006734**	**0.002944**	**0.007568**	**0.004616**	**0.005166**
Midilli et al.	a	0.998717	0.990274	0.998073	0.996553	0.995732	0.997813
	k	0.003900	0.003276	0.012181	0.005297	0.003729	0.003482
	n	1.072235	1.134644	0.972288	1.107234	1.271006	1.340274
	b	-0.000059	-0.000082	-0.000460	-0.000380	-0.000121	-0.000297
	R^2	0.999960	0.998238	0.999731	0.998804	0.998726	0.999149
	χ^2	0.000005	0.000227	0.000032	0.000223	0.000288	0.000315
	RMSE	0.001847	0.012722	0.004851	0.011139	0.012004	0.010253
Two-term exponential	a	-68.7268	-24.8089	1.055885	1.029677	1.868099	1.922440
	b	-0.0090	-0.0037	-0.010401	-0.008905	-0.008125	-0.008453
	c	69.7242	25.8144	-0.062844	-0.018581	-0.852491	-0.908642
	d	-0.009000	-0.003800	0.002326	0.007190	-0.004319	-0.003412
	R^2	0.999866	0.998121	0.999684	0.996763	0.996461	0.996137
	χ^2	0.000015	0.000242	0.000038	0.000604	0.000800	0.001431
	RMSE	0.003360	0.013139	0.005260	0.018325	0.020003	0.021839
Logarithmic	a	1.079914	1.112903	1.113792	1.223011	1.166145	1.301408
	k	0.005197	0.005698	0.009859	0.007089	0.010071	0.010223
	c	-0.071852	-0.104904	-0.122148	-0.218417	-0.149665	-0.287653
	R^2	0.999749	0.997672	0.999671	0.998634	0.995773	0.995714
	χ^2	0.000026	0.000272	0.000036	0.000213	0.000765	0.001058
	RMSE	0.004603	0.014622	0.005369	0.011905	0.021863	0.023003

CONCLUSION

In this study, oven drying and vacuum-assisted oven drying of blueberries were conducted, through which the effect of vacuum on the drying characteristics were investigated. Drying temperatures were set to 60, 70 and 80°C. The drying kinetic parameters of D_{eff} and E_a were calculated, and the drying curve data were fitted to twelve mostly used mathematical models present in the literature. The vacuum assistance was seen to decrease the drying times from 420, 390 and 210 minutes to 240, 210 and 150 minutes, for the temperatures of 60, 70 and 80°C, respectively. Considering the prominently reduced drying times with vacuum assistance, it can be said that the foresaid method offers economic benefits. The reduced drying times also decrease the amount of energy used for drying, hence being favorable in terms of environmental contribution. The drying rate curves showed that at the beginning of the drying (first 30 minutes), a rising-rate period was seen followed by a falling-rate period for all of the experiments. As the drying temperature increased, D_{eff} values also increased. Vacuum assistance was seen to increase both D_{eff} and E_a. D_{eff} values were calculated between 1.14×10^{-9}-2.83×10^{-9} m^2/s and 2.54×10^{-9}-4.33×10^{-9} m^2/s, for oven and vacuum-assisted oven drying, respectively. E_a for oven drying was calculated as 25.93 kJ/mol, and increased to 43.98 kJ/mol by the addition of vacuum. Yielding the highest R^2 values between 0.999968-0.999506 for oven drying and 0.999784-0.999243 for vacuum assisted oven drying, Alibas model was determined as the best model for both drying methods. For the future studies, different pretreatments can be applied before the drying process.

NOMENCLATURE

AOAC	Association of Official Analytical Chemists
D_{eff}	The effective diffusivity, m^2/s
DR	Drying rate, kg water/kg dry matter×min
E_a	Activation energy
L	Half-length of the sample, m
m_d	Mass of dry matter, g
m_w	Mass of water, g
M_0	Initial water content, kg water/kg dry matter
M_e	Water content at equilibrium, kg water/kg dry matter
M_t	Water content at time t, kg water/kg dry matter
M_f	Final water content, kg water/kg dry matter
MR	Moisture ratio, dimensionless
$MR_{exp,i}$	The experimental moisture ratio
$MR_{pre,i}$	The predicted moisture ratio
n	Number of models' constants
N	Number of data
t	Time

AUTHORSHIP CONTRIBUTIONS

Authors equally contributed to this work.

DATA AVAILABILITY STATEMENT

The authors confirm that the data that supports the findings of this study are available within the article. raw data that support the finding of this study are available from the corresponding author, upon reasonable request.

CONFLICT OF INTEREST

The author declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this article.

ETHICS

There are no ethical issues with the publication of this manuscript.

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