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Development of a New Model for Mass Transfer Kinetics of Petals of *Echium amoenum* Fisch. & C.A. Mey. under Fluidized Bed Conditions

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Summary

A new semi-theoretical thin-layer model of the fluidized bed drying of *Echium amoenum* Fisch. & C.A. Mey. petals has been developed. Experiments were conducted under different conditions: temperatures of 40, 50 and 60 °C, and air velocities of 0.5, 0.75 and 1.0 m/s, until the moisture content decreased to 0.04.–0.06 kg of water per kg of dry matter. Drying processes in the fluidized bed were completed in between 55 and 465 min, with minimum drying time required at the maximum temperature of 60 °C and air velocity of 1 m/s. The comparison of the new model developed here with sixteen previously published theoretical, semi-empirical or empirical thin-layer drying equations shows that using the new model the highest coefficient of correlation, the lowest reduced chi-square and root mean square error were obtained. The highest retention of total phenolic compounds in *E. amoenum* petals was achieved when drying at 60 °C and 1.0 m/s.

Key words: new thin-layer drying model, fluidized bed dryer, *Echium amoenum* petals, mass transfer kinetics, total phenolic content, optimum drying conditions

Introduction

Growing at an altitude ranging from 60 to 2200 m (*1*), *Echium amoenum* Fisch. & C.A. Mey. (Boraginaceae) is a biennial or perennial herb indigenous to the narrow zone of Northern Iran and Caucasus area. The dried violet-blue petals of *E. amoenum*, also known in the Iranian traditional medicine as Gol-e-Gavzaban, have long been used as a tonic, tranquillizer, diaphoretic and a remedy for cough, sore throat and pneumonia (*2,3*). The quality of freshly harvested *E. amoenum* petals deteriorates rapidly if they are not dried immediately. In Iran *E. amoenum* petals are traditionally sun-dried and even though this process is economical, the mechanical drying would be much more time-efficient, prevent losses and produce superior products compared with sun drying (*4*).

The fluidized bed is one of the most preferred techniques with a wide range of applications in different industries for drying fertilizers, chemicals, pharmaceuticals and minerals, for use as simple absorbers, in wastewater systems and complex reactors. New designs enable the application of fluidized bed for drying coarse material that does not fluidize easily. Compared with other drying methods, fluidized beds offer some advantages including a higher heat capacity, improved rates of heat and mass transfer between the phases, and ease of handling and transportation of fluidized solids (*5*).

The drying kinetics is a complex phenomenon requiring a simple method in order to predict the drying behaviour and optimize the drying parameters. Thus, thin-layer drying equations have been used for prediction of drying time and for generating drying curves (*6*). While the thin-layer drying curves of fruits and vegetables are usually modelled using empirical, semi-empirical or analytical equations, some of the popular mathematical models

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such as the logarithmic equation are used for pumpkin, strawberry and mushroom (*7–9*), Page equation for green chilli, flax fibre, young coconut, sweet potato, carrot and kiwifruit (*10–15*), Henderson-Pabis equation for fruits (*16*), Thompson equation for green peas (*17*), single-term exponential model for onion slices (*18*) and Midilli equation for fruits and vegetables (*19,20*).

The present study aims to develop a new empirical model that would be more compatible with the drying kinetics. Furthermore, the new empirical model is compared with sixteen empirical or semi-empirical models published in the literature and commonly used for fruits and vegetables. Although the decoct of *E. amoenum* dry petals is used widely in the folk medicine, very few studies have focused on their drying (*21*); therefore, this research has been conducted in an attempt to determine the effect of drying conditions on total phenolic content.

Materials and Methods

Fluidized bed dryer

A laboratory-scale fluidized bed dryer (Fig. 1) constructed at the Department of Agricultural Machinery Engineering of Azadshahr University, Iran, was used to conduct drying experiments. It consisted of an air-heating device, chamber, and control systems for air and temperature. The fluidized bed chamber, made of stainless steel with inside dimensions of 250 mm×250 mm×300 mm, is connected with a fan, a duct made of stainless steel to supply air, while the distributor plate, with thickness of 1 mm and holes of 3 mm in diameter, was tightly fixed to the bottom of the chamber. The fluidizing air is supplied by a 1.5-kW blower, while an anemometer is used to regulate the air velocity manually. The heating unit consists of three fin heaters each of 800 W using thermostat-type temperature controls in order to adjust the desired drying temperature.

Drying experiments

In order to determine the drying kinetics, the *Echium amoenum* petals were dried in a fluidized bed dryer at 40, 50 and 60 °C and three different air velocities (0.5, 0.75 and 1 m/s). Collected from Ali-Abad (Golestan Province, Iran), fresh *E. amoenum* petals used in the drying experiments were stored at (4.0±0.5) °C before the beginning of the drying process. Their initial moisture content of about 8.67 to 10.29 kg of water per kg of dry matter was determined by oven drying at 105 °C according to the Polish Standard PN-R-87019:1991 (*22*). Nearly 1.4 kg/m^2 of *E. amoenum* petals was uniformly spread in a thin layer on a perforated stainless steel tray for drying, and moisture loss was recorded at 5-minute intervals by a digital precision balance of 0.01 g accuracy (A&D-FX-1200i; A&D Company, Ltd, Tokyo, Japan). The experiment was repeated three times, and each time the drying continued until there was not any change in the moisture content observed.

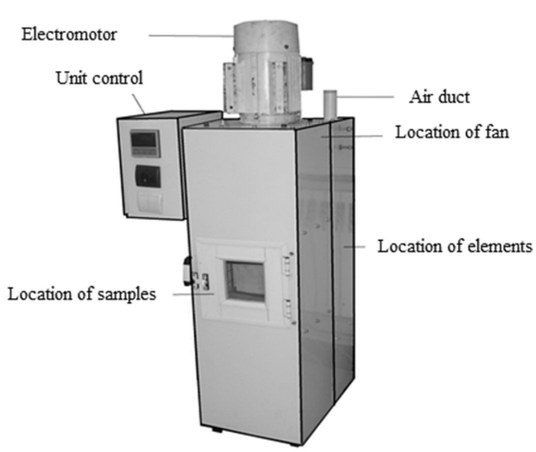

Fig. 1. Fluidized bed dryer

Mathematical modelling of the drying curve

The proposed equation for the modelling of the drying curve is as follows:

$$\mathrm{MR}=a\exp\left(-k_1t^{n}\right)+b\exp\left(-k_2t\right)+c \qquad /1/$$

where MR is the moisture ratio, which can be calculated as follows:

$$\mathrm{MR}=\frac{\mathrm{M_t}-\mathrm{M_e}}{\mathrm{M_0}-\mathrm{M_e}} \qquad /2/$$

where M_t, M_0 and M_e are the moisture content at time *t*, the initial and equilibrium points, respectively, expressed in percentage on dry matter basis. During thin-layer drying of *E. amoenum* petals in fluidized bed dryer, the samples were not exposed to uniform relative humidity and temperature continuously; therefore, the moisture ratio was simplified according to Doymaz (*14*) and Pala *et al.* (*23*) to:

$$\mathrm{MR}=\frac{\mathrm{M_t}}{\mathrm{M_e}} \qquad /3/$$

In order to select a suitable model to describe the drying process of *E. amoenum* petals, the drying curves were fitted with the newly proposed model and sixteen thin-layer drying equations from the literature. The evaluated moisture ratio models are shown in Table 1 (*13,16,24–37*).

Nonlinear regression analyses of these equations were applied using MATLAB v. 7.8.0.347 (MATLAB Inc., Natick, MA, USA) in order to estimate the parameters *k*, k_0, k_1, k_2, *a*, a_0, *b*, *c*, *g*, *h*, *L* and *n* of empirical and semi-empirical equations in Table 1. The coefficient of correlation, R^2, is one of the main criteria for selecting the best model, and the appropriate fitting status was determined by various statistical parameters such as reduced chi-square (χ^2) and root mean square error (RMSE) values. Higher values of R^2 with lower χ^2 and RMSE values lead to a well-closed and suitable fitting. The following equations were used to determine the aforementioned parameters:

$$R^2=\left[\frac{N\sum_{i=1}^{N}\mathrm{MR}_{\mathrm{exp},i}\cdot\mathrm{MR}_{\mathrm{pre},i}-\sum_{i=1}^{N}\mathrm{MR}_{\mathrm{exp},i}\cdot\sum_{i=1}^{N}\mathrm{MR}_{\mathrm{pre},i}}{\left(\sqrt{N\left(\sum_{i=1}^{N}\mathrm{MR}^2_{\mathrm{exp},i}\right)\left(\sum_{i=1}^{N}\mathrm{MR}_{\mathrm{exp},i}\right)^2}\cdot\sqrt{N\left(\sum_{i=1}^{N}\mathrm{MR}^2_{\mathrm{pre},i}\right)\left(\sum_{i=1}^{N}\mathrm{MR}_{\mathrm{pre},i}\right)^2}\right)}\right]^2 \qquad /4/$$

Table 1. Mathematical thin-layer drying models

No.	Model	Equation	Reference
1	Page	$MR=\exp(-kt^n)$	(*24*)
2	Modified Page	$MR=a\exp(-(kt)^n)$	(*25*)
3	Modified Page equation II	$MR=\exp[-k(t/L^2)^n]$	(*26*)
4	Modified Henderson-Pabis	$MR=a\exp(-kt)+b\exp(-gt)+c\exp(-ht)$	(*16*)
5	Two-term	$MR=a\exp(-k_0t)+b\exp(-k_1t)$	(*27*)
6	Two-term exponential	$MR=a\exp(-kt)+(1-a)\exp(-kat)$	(*28*)
7	Thompson	$t=a\,ln(MR)+b[ln(MR)]^2$	(*29*)
8	Weibull distribution	$MR=a-b\exp(-kt^n)$	(*30*)
9	Aghbashlo *et al.*	$MR=\exp(-k_1t)/(1+k_2t)$	(*31*)
10	Logistic	$MR=a_0/(1+a\exp(kt))$	(*32*)
11	Simplified Fick's diffusion	$MR=a\exp[-c(t/L^2)]$	(*13*)
12	Demir *et al.*	$MR=a\exp(-kt)^n+c$	(*33*)
13	Alibas	$MR=a\exp(-kt^n+bt)+g$	(*34*)
14	Jena and Das	$MR=a\exp(-kt+b\sqrt{t})+c$	(*35*)
15	Verma *et al.*	$MR=a\exp(-kt)+(1-a)\exp(-gt)$	(*36*)
16	Parabolic	$MR=at^2+bt+c$	(*37*)
17	New model	$MR=a\exp(-k_1t^n)+b\exp(-k_2t)+c$	this paper

MR=moisture ratio, t=time of drying

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

$$c^2=\frac{\left[\sum_{i=1}^{N}\mathrm{MR}_{\mathrm{exp,i}}\sum_{i=1}^{N}\mathrm{MR}_{\mathrm{pre,i}}\right]^2}{N-z} \quad /6/$$

where MR_{exp}, MR_{pre}, N and z are the experimental and predicted moisture ratios, number of observations and number of drying constants, respectively.

Total phenolic content analysis

Total polyphenol content was estimated using Folin--Ciocalteu's assay (*38,39*). A known concentration of *E. amoenum* petal extract (10 mg/mL) was mixed with 1.0 mL of Folin-Ciocalteu's reagent, 0.8 mL of Na_2CO_3 was added and the mixture volume was then increased to 10 mL, using a water/methanol mixture (4:6). Absorbance was read at 740 nm after 30 min using a spectrophotometer (type 108; Systronics, Ahmedabad, Gujarat, India). Using tannic acid (0–800 mg/L) to produce a standard calibration curve, the total phenolic content was expressed in mg of tannic acid equivalents (TAE) per 100 g of sample (*40*).

Statistical analysis

Two full-factorial experimental designs were used in this research, and ANOVA was performed using PASW (Predictive Analytics Software) statistics software v. 18.0 (SPSS Inc., Chicago, IL, USA) to determine the influence of the contribution of the independent variables (temperature and air velocity) on both the total phenolic content loss and drying time using a significance level of 1 %. All measurements were done in triplicate, the data were presented as mean values and compared with others using Duncan's multiple range tests. The linear or non-linear regression procedures were applied to determine the relationship of independent variables with total phenolic content loss and drying time, and regression coefficients (R^2) were given to assess the accuracy of the obtained relationships.

Results and Discussion

Drying curves

An average initial moisture content of *E. amoenum* petals was from 8.67 to 10.29 kg of water per kg of dry matter, which was reduced to 0.04 to 0.06 kg/kg at 40, 50 and 60 °C and drying air velocities of 0.5, 0.75 and 1 m/s, using a fluidized bed dryer. The time to reduce the moisture ratio to any given level depended on the drying conditions, thus it took 55–465 min in the fluidized bed dryer, and about 2–3 days when sun drying in the open air in May and June of 2014.

Different values of moisture ratio as a function of time required for *E. amoneum* petals to be dried depend on the different drying conditions in fluidized bed dryer (Figs. 2 and 3). The influence of the drying temperature (40, 50 and 60 °C) on moisture ratio is shown in Fig. 2. It has been shown that in the temperature range of this experiment, higher temperatures lead to the faster drying rate and result in shorter drying time, which is due to the increased heat transfer gradient between the air and the petals, which facilitates water evaporation from the petals.

Moisture content reduction was faster at the beginning of the process than at the end, due to the slow diffusion process. This can be explained by the fact that the petals contained a large quantity of bulk water in the beginning, which was easily transferred to the surface and

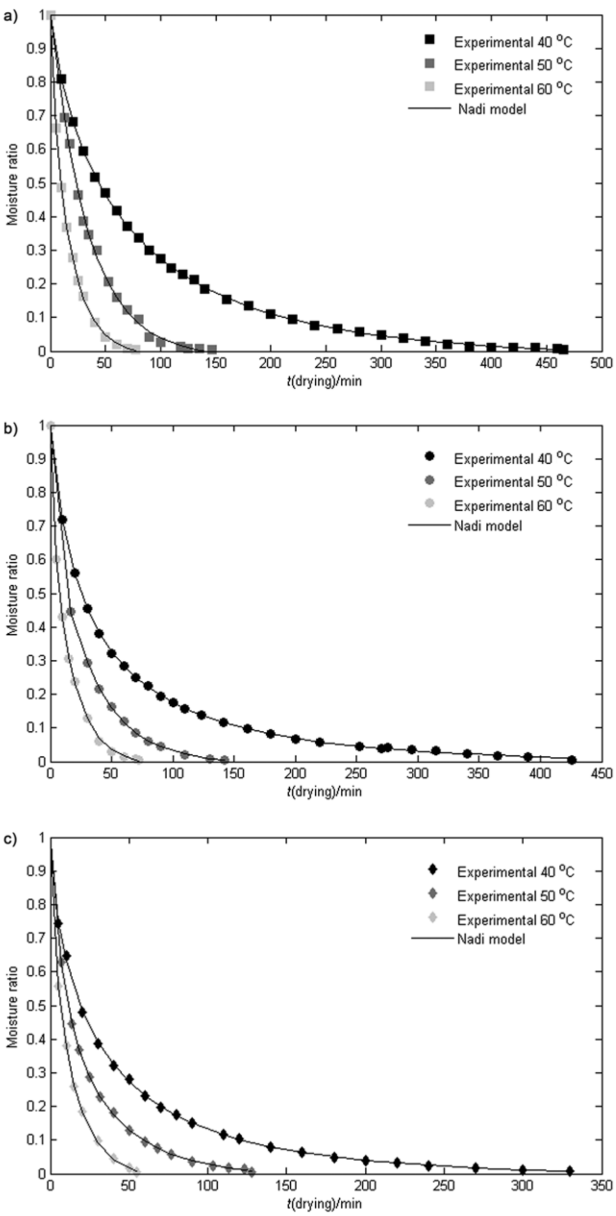

Fig. 2. Comparison of experimental with predicted curves of moisture ratio *vs.* time in a fluidized bed dryer using the new equation (model no. 17, Table 1) at air velocity of: a) 0.5, b) 0.75 and c) 1 m/s

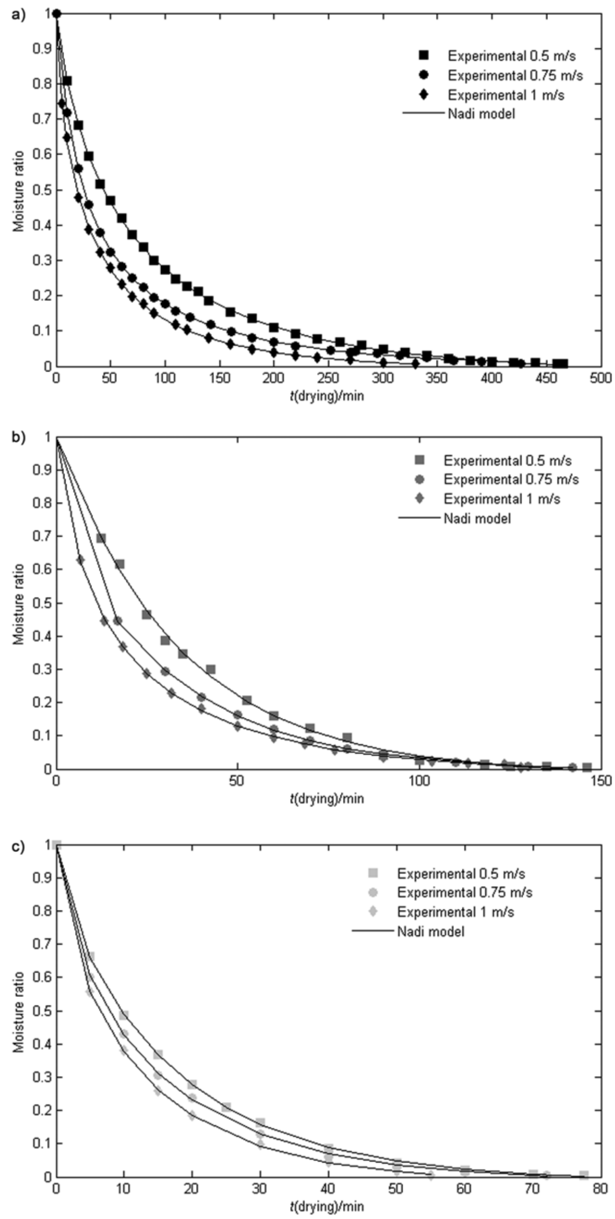

Fig. 3. Comparison of experimental with predicted curves of moisture ratio *vs.* time in fluidized bed dryer using the new equation (model no. 17, Table 1) at temperatures of: a) 40, b) 50 and c) 60 °C

evaporated. As the drying time increased, the bulk water content was significantly reduced, while bound water was more difficult to transfer and therefore the drying process slowed down. This can even lead to the 'hard shell' effect, causing a significant decrease in the diffusion and drying rates.

With an increase in the air velocity, the drying rate of *E. amoenum* petals also increased (Fig. 3). The values of moisture ratio between the experimental drying curves in Fig. 3 are smaller than those in Fig. 2, which means that increasing the velocity of the hot air cannot shorten the drying time notably; on the contrary, it may only result in wasting energy. The results proved that the process of drying *E. amoenum* petals was controlled by internal moisture diffusion. As the moisture evaporation rate on the surface of *E. amoenum* petals was faster than the moisture diffusion within them, internal moisture did not transfer to the surface to evaporate, which is the main reason why the hot air velocity had less obvious effects on the moisture ratio and the drying speed. In the selected temperature range at a specific drying time the difference in the moisture ratio was smaller at a higher (1.0 m/s) than at a lower air velocity (0.5 m/s).

Mathematical modelling

It has been observed that the Page model (model no. 1 in Table 1) is the best among the different models including the models of Lewis, Henderson-Pabis, logarithmic, Wang and Singh, diffusion approximation and Midilli–Kucuk for thin layer drying of *E. amoenum* petals in a fluidized bed dryer (*21*).

In this study, seven additional models have been evaluated including a new model and sixteen thin-layer drying models defined by various researchers presented in Table 1. The coefficient of correlation (R^2), root mean square error (RMSE) and chi-square (χ^2) obtained in the thin-layer fluidized bed drying are shown in Tables 2–4. A special empha-

Table 2. Statistical results of the different equations for thin-layer fluidized bed drying of *Echium amoenum* petals at 40 °C

No.	*v*(air)/(m/s)								
	0.5			0.75			1		
	R^2	RMSE	χ^2	R^2	RMSE	χ^2	R^2	RMSE	χ^2
1	0.9996	0.0089	0.00000	0.9989	0.0082	0.00000	0.9996	0.0053	0.00000
2	0.2608	0.2967	0.97700	0.4693	0.2341	0.06170	0.4377	0.2638	0.08060
3	0.9997	0.0056	0.00003	0.9993	0.0082	0.000075	0.9998	0.0052	0.000030
4	0.6425	0.2070	0.05270	0.6580	0.1829	0.04300	0.4021	0.2417	0.93460
5	0.9997	0.0046	0.00057	0.9996	0.0047	0.00056	0.9989	0.0094	0.00023
6	0.9984	0.0140	0.00021	0.9904	0.0306	0.00100	0.9884	0.0370	0.00150
7	0.9968	847.05	765320	0.9911	1196.9	1547.10	0.9953	707.06	547540
8	0.8526	0.1329	0.02020	0.9995	0.0070	0.00006	0.9028	0.1073	0.0139
9	0.9952	0.0182	0.00965	0.9955	0.1628	0.00716	0.9895	0.0280	0.01641
10	0.6218	0.2129	0.05000	0.6790	0.1772	0.03530	0.6568	0.2016	0.04670
11	0.2608	0.2976	0.09770	0.3789	0.2464	0.06830	0.3672	0.2737	0.08620
12	0.9999	0.0039	0.00002	0.6790	0.1772	0.03680	0.7117	0.1847	0.04130
13	0.6218	0.2129	0.05370	0.9092	0.0942	0.01090	0.6568	0.2016	0.05190
14	0.6246	0.2121	0.05140	0.5328	0.2137	0.05360	0.7575	0.1694	0.03480
15	0.9998	0.0043	0.00002	0.9998	0.0047	0.00002	0.9994	0.0088	0.00009
16	0.9552	0.0732	0.00590	0.9008	0.0985	0.01090	0.9061	0.1055	0.01280
17	**0.9999**	**0.0031**	**0.00001**	**0.9999**	**0.0025**	**0.00000**	**0.9998**	**0.0046**	**0.00003**

R^2=coefficient of correlation, RMSE=root mean square error, χ^2=chi-square

Table 3. Statistical results of the different equations for thin-layer fluidized bed drying of *Echium amoenum* petals at 50 °C

No.	*v*(air)/(m/s)								
	0.5			0.75			1		
	R^2	RMSE	χ^2	R^2	RMSE	χ^2	R^2	RMSE	χ^2
1	0.9956	0.0406	0.00190	0.9996	0.0077	0.00000	0.9997	0.0049	0.00000
2	0.4392	0.2890	0.10150	0.7223	0.179	0.04270	0.5176	0.2414	0.07170
3	0.9990	0.0121	0.00020	0.9996	0.0067	0.00006	0.9998	0.0050	0.00003
4	0.7501	0.1929	0.05750	0.9869	0.0389	0.00300	0.5176	0.2414	0.09320
5	0.9988	0.0028	0.00067	0.9999	0.0035	0.00051	0.9999	0.0024	0.00007
6	0.9987	0. 0140	0.00022	0.9995	0.0073	0.00006	0.9971	0.0188	0.00040
7	0.9977	229.97	59937.0	0.9988	165.67	329360	0.9946	319.33	116540
8	0.8627	0.1430	0.02670	0.9999	0.0030	0.00001	0.8456	0.1365	0.02490
9	0.9984	0.0118	0.00223	0.9980	0.0138	0.00171	0.9961	0.01761	0.00433
10	0.6966	0.2126	0.05490	0.8624	0.1260	0.02120	0.7494	0.1739	0.03720
11	0.3949	0.3002	0.10950	0.7223	0.1790	0.04270	0.5176	0.2414	0.07170
12	0.9992	0.0109	0.00016	0.9999	0.0030	0.00001	0.9999	0.0038	0.00001
13	0.6966	0.2126	0.06400	0.8624	0.1260	0.02720	0.7494	0.1739	0.04400
14	0.6966	0.2126	0.05910	0.8624	0.1260	0.02380	0.9997	0.0060	0.00004
15	0.9992	0.0109	0.00014	0.9999	0.0029	0.00001	1.0000	0.0019	0.00000
16	0.9855	0.0464	0.0026	0.9427	0.0813	0.00880	0.9368	0.0874	0.00940
17	**0.9992**	**0.0108**	**0.00018**	**1.0000**	**0.0019**	**0.00000**	**1.0000**	**0.0024**	**0.00000**

For abbreviations see Table 2

Table 4. Statistical results of the different equations for thin-layer fluidized bed drying of *Echium amoenum* petals at 60 °C

No.	*v*(air)/(m/s)								
	0.5			0.75			1		
	R^2	RMSE	χ^2	R^2	RMSE	χ^2	R^2	RMSE	χ^2
1	0.9993	0.0083	0.00001	0.9991	0.0097	0.00010	0.9994	0.0079	0.00001
2	0.5081	0.2480	0.10753	0.5821	0.2555	0.08970	0.6369	0.2517	0.09500
3	0.9995	0.0089	0.00010	0.9994	0.0097	0.00013	0.9996	0.0079	0.00009
4	0.9979	0.0183	0.00067	0.9999	0.0048	0.00005	0.6805	0.3685	0.27161
5	0.9997	0.0063	0.00032	0.9998	0.0052	0.00022	0.9998	0.0058	0.00016
6	0.9998	0.0050	0.00003	0.9995	0.0085	0.00008	0.9995	0.0091	0.00010
7	0.9984	100.85	122062	0.9993	65.002	5164.20	0.9952	130.296	21828.0
8	0.9814	0.0553	0.00461	0.9798	0.0971	0.01480	0.8954	0.1351	0.03280
9	0.9973	0.0167	0.00279	0.9966	0.0160	0.00368	0.9971	0.01881	0.00248
10	0.7583	0.1991	0.05283	0.7751	0.1874	0.04830	0.8282	0.1731	0.04500
11	0.5081	0.2840	0.10751	0.5821	0.2555	0.08970	0.6369	0.2517	0.09500
12	0.9999	0.0040	0.00002	0.9998	0.0058	0.00005	1.0000	0.0018	0.00001
13	0.7583	0.1991	0.06792	0.7751	0.1874	0.06440	0.8282	0.1731	0.06740
14	0.9998	0.0043	0.00002	0.9998	0.0047	0.00002	0.9994	0.0088	0.00009
15	0.9999	0.0049	0.00003	0.9999	0.0048	0.00003	0.9999	0.0050	0.00004
16	0.9719	0.0679	0.00611	0.9583	0.0807	0.00890	0.9634	0.0799	0.00960
17	**0.9999**	**0.0032**	**0.00002**	**0.9999**	**0.0040**	**0.00003**	**1.0000**	**0.0023**	**0.00001**

For abbreviations see Table 2

sis is put on the R^2 as one of the primary criteria for selecting the best model for defining the hot-air drying curves of the petals. Thin-layer drying model in which the R^2 is mostly close to 1.0000 and RMSE and χ^2 values are mostly near 0.0000 was the optimum model. The new model proposed here has the highest R^2 and the lowest RMSE and χ^2

Table 5. Coefficients (see Table 1) obtained from different thin-layer drying models at 40 °C

No.	*v*(air)/(m/s)		
	0.5	0.75	1
1	k=0.9366 n=0.7310	k=1.5023 n=0.9156	k=3.4225 n=0.8725
2	a=1.0000 k=−8.2373 n=5.6308	a=0.2868 k=0.0861 n=0.0000	a=0.3071 k=0.0848 n=0.0000
3	k=0.0461 L=−8.9575 n=0.7739	k=0.1112 L=−9.9878 n=0.6703	k=0.1416 L=−9.9834 n=0.6507
4	a=0.0932 k=0.0001 b=0.0932 g=0.0001 c=0.0932 h=0.0001	a=0.0951 k=0.0001 b=0.0946 g=0.0001 c=0.0950 h=0.0001	a=0.8170 k=0.357 b=0.0027 g=0.7533 c=0.2954 h=0.1145
5	a=0.6974 k_0=−0.0001 b=0.2999 k_1=−0.0009	a=0.5485 k_0=−0.0009 b=0.4465 k_1=−0.0001	a=0.4305 k_0=−0.0018 b=0.5604 k_1=−0.0002
6	a=0.2172 k=0.0008	a=0.2434 k=0.0011	a=0.2124 k=0.0017
7	a=−5205.3 b=0.0815	a=−3067.1 b=0.5173	a=−2528.7 b=0.2978
8	a=13.5519 b=12.9917 k=−0.0008 n=0.3831	a=0.0111 b=−0.9945 k=0.0045 n=0.6900	a=10.5990 b=9.9447 k=−0.0012 n=0.4091
9	k_1=0.0003 k_2=4.187·10^{-5}	k_1=0.0005 k_2=0.0001	k_1=0.0007 k_2=0.0001
10	a_0=0.2113 a=−0.7887 k=−0.1827	a_0=0.1745 a=−0.8255 k=−0.0714	a_0=0.1893 a=−0.8107 k=−6.5737
11	a=1.0000 c=0.0001 L=0.0001	a=1.0000 c=0.0000 L=0.0000	a=1.0000 c=0.1000 L=0.1000
12	a=1.0170 k=0.0002 n=0.7475 c=−0.0132	a=0.8255 k=9.8271 n=1.8407 c=0.1745	a=0.8616 k=13.0200 n=0.0791 c=0.1166
13	a=0.7887 k=10.0007 n=0.4276 b=0.0100 g=0.2113	a=0.8264 k=−0.0025 n=0.0000 b=−0.0006 g=0.1734	a=0.8107 k=0.0000 n=0.0000 b=−0.4936 g=0.1893
14	a=0.7888 k=−0.0010 b=−0.2138 c=0.2116	a=0.0695 k=0.0003 b=0.0105 c=0.1945	a=0.3666 k=0.0001 b=−0.0166 c=0.1084
15	a=0.2999 k=0.0009 g=0.0002	a=0.5488 k=0.009 g=0.002	a=0.4344 k=0.0019 g=0.002
16	a=0.000 b=−0.0001 c=0.7363	a=0.0000 b=−0.0001 c=0.6427	a=0.0000 b=−0.0001 c=0.6595
17	**a=1.0715 k_1=0.0029 n=0.7000 b=−0.0509 k_2=−0.0596 c=−0.0207**	**a=1.2861 k_1=0.0188 n=0.5332 b=−0.2780 k_2=−0.0790 c=−0.0078**	**a=1.0423 k_1=0.0095 n=0.6155 b=−0.0333 k_2=−0.1027 c=−0.0089**

values among all drying tests (Tables 2–4), so it can fit suitably to the data, compared to the other models investigated in this paper and our previous study (*21*).

The fluidized bed drying constants and the coefficients of the thin-layer drying models are shown in Tables 5–7.

Table 6. Coefficients (see Table 1) obtained from different thin-layer drying models at 50 °C

No.	v(air)/(m/s)		
	0.5	0.75	1
1	k=1.2163 n=0.6487	a=2.1620 n=0.7989	a=3.6850 n=0.8150
2	a=0.3096 k=0.08557 n=0.0000	a=1.0000 k=−9.7343 n=1.7047	a=1.0000 k=−9.7326 n=1.7020
3	k=0.0004 L=0.0100 n=1.0475	k=0.0021 L=0.0100 n=0.8018	k=0.0037 L=0.1000 n=0.7346
4	a=0.1382 k=0.0002 b=0.1341 g=0.0002 c=0.1408 h=0.0002	a=0.2893 k=0.0004 b=0.5509 g=0.0008 c=0.0277 h=0.0007	a=0.3333 k=0.1000 b=0.3333 g=0.1000 c=0.3333 h=0.1000
5	a=0.1425 k_0=0.0001 b=0.1549 k_1=0.0001	a=0.2517 k_0=−0.00042 b=0.7483 k_1=−0.00052	a=0.6180 k_0=−0.00052 b=0.3892 k_1=−0.00273
6	a=0.0033 k=0.1520	a=0.2496 k=0.0021	a=0.2410 k=0.0027
7	a=−2099.1 b=−0.0959	a=−1668.9 b=−0.0011	a=−1418.4 b=0.0447
8	a=9.1412 b=8.5296 k=−0.0014 n=0.4253	a=−0.0232 b=−1.0230 k=0.0045 n=0.7429	a=17.0276 b=16.4812 k=−0.0009 n=0.3968
9	k_1=0.0005 k_2=$-2.009\cdot10^{-5}$	k_1=0.0008 k_2=$9.049\cdot10^{-5}$	k_1=0.0011 k_2=0.0002
10	a_0=0.2185 a=−0.7815 k=−0.0308	a_0=0.1328 a=−0.8672 k=−0.2437	a_0=0.1728 a=−0.2493 k=−0.0586
11	a=1.0000 c=0.0100 L=0.0100	a=1.0000 c=0.1000 L=0.1000	a=1.0000 c=0.0100 L=0.0100
12	a=1.0162 k=0.0005 n=1.0102 c=−0.0152	a=1.0230 k=0.0007 n=0.7429 c=−0.0232	a=1.0124 k=0.0009 n=0.7110 c=−0.0112
13	a=0.7815 k=0.0000 n=0.0000 b=−0.4872 g=0.2185	a=0.8672 k=0.0003 n=0.0001 b=−0.3211 g=0.1328	a=0.8272 k=0.0000 n=0.0000 b=−0.3210 g=0.1728
14	a=0.7815 k=0.3614 b=−9.9866 c=0.2185	a=0.8672 k=0.0609 b=−0.5979 c=0.1328	a=1.0015 k=0.0004 b=−0.0170 c=0.0006
15	a=0.0686 k=0.00001 g=0.0005	a=−0.7503 k=0.0005 g=0.0044	a=0.6109 k=0.0005 g=0.0027
16	a=0.0000 b=−0.0003 c=0.8858	a=0.0000 b=−0.0003 c=0.8275	a=0.0000 b=−0.0003 c=0.7505
17	**a=1.0578 k_1=0.0007 n=0.9656 b=−0.0390 k_2=−0.4695 c=−0.0188**	**a=0.7753 k_1=0.0007 n=0.9638 b=0.2322 k_2=−0.0790 c=−0.0073**	**a=1.1349 k_1=0.0128 n=0.6322 b=−0.1127 k_2=−0.0676 c=−0.0218**

Table 7. Coefficients (see Table 1) obtained from different thin-layer drying models at 60 °C

No.	v(air)/(m/s)		
	0.5	0.75	1
1	k=1.4571 n=0.6487	k=2.5977 n=0.6392	k=5.334 n=0.7055
2	a=1.0000 k=−9.7044 n=1.8457	a=1.0000 k=−9.6902 n=1.9356	a=1.0000 k=−9.6711 n=2.0316
3	k=0.0021 L=0.1000 n=0.8719	k=0.0033 L=0.1000 n=0.8169	k=0.0043 L=0.1000 n=0.8007
4	a=0.3216 k=0.0011 b=0.3267 g=0.0011 c=0.3205 h=0.0011	a=0.1808 k=0.3209 b=0.1979 g=0.0011 c=0.6213 h=0.0011	a=0.8068 k=0.3592 b=0.005126 g=0.7523 c=0.2948 h=0.1145
5	a=0.8706 k_0=−0.00096 b=0.1294 k_1=−0.0086	a=0.2916 k_0=−0.0098 b=0.8084 k_1=−0.00105	a=0.7658 k_0=−0.001184 b=0.2342 k_1=−0.00795
6	a=0.1359 k=0.0070	a=0.1942 k=0.0055	a=0.2177 k=0.0057
7	a=1009.4 b=−0.0254	a=−828.7881 b=3.4995	a=−800.5050 b=−24.5709
8	a=10.3679 b=9.4232 k=−0.0051 n=0.3567	a=10.0469 b=15.0012 k=−0.0016 n=0.4636	a=31.6363 b=31.0013 k=−0.0007 n=0.4216
9	k_1=0.0012 k_2=$9.073\cdot10^{-5}$	k_1=0.0015 k_2=0.0002	k_1=0.0019 k_2=0.0002
10	a_0=0.2116 a=−0.7884 k=−0.1552	a_0=0.1820 a=−0.8180 k=−0.1558	a_0=0.1938 a=−0.8062 k=−0.1484
11	a=1.0000 c=0.1000 L=0.1000	a=1.0000 c=0.0000 L=0.0000	a=1.0000 c=0.1000 L=0.1000
12	a=1.0258 k=0.0011 n=0.8225 c=−0.0267	a=1.0235 k=0.0013 n=0.7660 c=−0.0246	a=1.0321 k=0.0015 n=0.7393 c=−0.0322
13	a=0.7884 k=0.0001 n=0.0000 b=−0.4859 g=0.2116	a=0.8180 k=0.0001 n=0.0001 b=−0.1890 g=0.1820	a=0.8062 k=0.0002 n=0.0001 b=−0.2915 g=0.1938
14	a=0.7884 k=0.3311 b=0.1507 c=0.2116	a=0.8180 k=0.0598 b=−9.9964 c=0.1820	a=0.8062 k=0.3000 b=−2.4331 c=0.1938
15	a=0.1302 k=0.0085 g=0.001	a=0.1767 k=0.1572 g=0.0011	a=0.1976 k=0.1572 g=0.0012
16	a=0.0000 b=−0.0005 c=0.8454	a=0.0000 b=−0.0005 c=0.8191	a=0.0000 b=−0.0007 c=0.8517
17	**a=0.9348 k_1=0.0017 n=0.9228 k_2=0.0803 b=−0.4680 c=−0.0150**	**a=0.8588 k_1=0.0016 n=0.9434 b=0.1487 k_2=−0.1663 c=−0.0075**	**a=0.9229 k_1=0.0040 n=0.8348 b=0.0962 k_2=−0.2602 c=−0.0191**

Fig. 4 shows the predicted moisture ratio of the new model compared to the experimental results. The predicted moisture values distributed along the straight line, with a slope of 45°, demonstrate the appropriateness of the selected model regarding the thin-layer drying behaviour of *E. amoenum* petals.

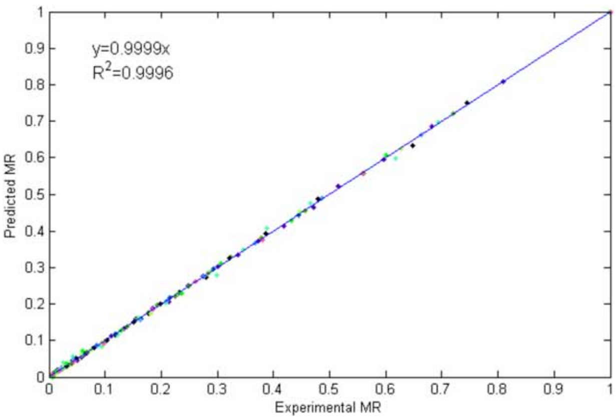

Fig. 4. Experimental *vs.* predicted moisture ratio (MR)

The effect of drying conditions on total phenolic content

Several scientific works have focused on the study of the effects of drying process on vegetable phenolic compounds and concluded that it can vary from having no effect at all (*41*), to causing significant losses (*42–46*) or enhancement of total phenolic content (TPC) (*47–49*). Therefore, TPC was evaluated in dried *E. amoenum* petals to obtain the percentage losses, and thus determine the effect of drying conditions on TPC change.

The results of the statistical analysis of variance (ANOVA) and Duncan's test of the data obtained from the experiment are presented in Table 8, and they show that there are significant differences among the TPC values ($p<0.01$), and also that the effect of drying air velocity and drying temperature on TPC losses is highly significant ($p<0.01$) in a way that at 60 °C, the highest drying temperature, degradation of total phenolics is the slowest. High drying temperatures could be the result of inactivation of enzymes responsible for the oxidation of polyphenols such as polyphenol oxidases and peroxidases, present in plant materials (*43,49*), while the formation of phenolic compounds at high temperatures might be caused by the availability of phenolic precursor molecules through nonenzymatic interconversion between phenolic molecules (*50*), which is probably due to high convective forces acting at the air–solid interface, and retarding heat diffusion into the petals. Being localised in hydrophilic regions of cells such as vacuoles and apoplasts or as other soluble phenols in the cytoplasm and in the cell nuclei (*51*), the phenolic glycosides are protected from heat by the cell wall material. Internal resistance to heat diffusion is therefore an important parameter to be considered that affects the quality of the product during heat treatment.

Similarly, maximum reduction in TPC was seen in onion dried at 50 °C, whereas a lower reduction was seen when drying at 70 °C (*52*). According to Gupta *et al.* (*49*) and Garau *et al.* (*53*), who concluded that longer drying times would result in higher reduction of TPC in orange by-products, important losses of TPC at lower drying temperature could also be attributed to the longer duration of drying.

It has been reported by Martín-Cabrejas *et al.* (*54*) and Mrad *et al.* (*55*) that TPC reduction might also be caused by the binding of polyphenols with other compounds or by the modification of their chemical structure following heat treatment, which would prevent their extraction and determination by the adopted methods.

Table 8. Total phenolic content (TPC) of fresh and dried *Echium amoenum* petals under different drying conditions

Air drying conditions		w(TPC)	TPC loss
v/(m/s)	Temperature/°C	mg/g dry matter	%
Fresh	–	0.81	–
0.5	40	$(0.322\pm0.002)^{f}$	60.49
	50	$(0.427\pm0.003)^{e}$	46.91
	60	$(0.563\pm0.005)^{b}$	30.86
0.75	40	$(0.444\pm0.001)^{de}$	45.68
	50	$(0.506\pm0.004)^{c}$	37.04
	60	$(0.614\pm0.007)^{a}$	24.69
1	40	$(0.441\pm0.002)^{ed}$	45.68
	50	$(0.461\pm0.003)^{d}$	43.21
	60	$(0.622\pm0.009)^{a}$	23.46

Mean values with different letters indicate significant differences ($\alpha=0.01$) at different drying conditions according to the Duncan's multiple range test

The following equation was derived from the experimental data in order to express the relationship between the TPC and the independent variables. Statistical results show that 97.7 % of TPC variability can be explained by this equation, with independent and identical distribution:

$$\mathrm{TPC}=0.143-0.02197T+1.609v+ \\ +0.0003633T^2-0.006T\cdot v-0.7787v^2 \qquad /7/$$

where T and v are drying temperature and air velocity, respectively.

Effect of drying conditions on drying time

Drying time is the time required to reduce the moisture content of *E. amoenum* petals to 5 % on dry mass basis. The ANOVA of drying time with respect to temperature and air velocity was analyzed (Table 9) and the F-values of temperature and air velocities were significant ($p<0.05$).

The results of modelling of the interaction effects of air velocity and temperature on drying time of the samples are shown in Fig. 5. It can be seen that the maximum drying time (465 min) was exhausted at the lowest temperature and air velocity (40 °C and 0.5 m/s, respectively) and the minimum drying time (55 min) was spent at the highest temperature and air velocity (60 °C and of 1.0

Table 9. ANOVA for drying time of *Echium amoenum* petals

Source of variation	SS	df	MS	F
Temperature	373851.556	2	4373.788	562.343*
Air velocity	8747.556	2	186925.778	24033.314*
Error		18		
Total		27		

*Level of significance $p<0.05$

SS=sum of squares, df=degree of freedom, MS=mean sum of squares

m/s, respectively). For comparison, the longest drying time is 8.29 times longer than the shortest. While drying at higher temperature and higher velocity of air circulation intensifies the drying rate of the *E. amoenum* petals and leads to a shorter drying time, it has also been observed that within certain ranges of air velocity (0.5–1.0 m/s) and temperature (40–60 °C), the temperature had stronger effect than air velocity, which was also observed by Kumar *et al.* (*56*) during the thin-layer drying of carrot pomace, rice flour and production of pulse powder.

Drying time is significantly affected by the linear terms of all the independent variables. Both drying temperature and air velocity have a negative linear effect on total drying time (Fig. 5). The following equation was obtained from the experimental data to express the relationship between the total drying time and process variables.

$$DT=3971-125.4T-610v+1.008T^2+10T\cdot v \qquad /8/$$

The statistical results show that 99.2 % of drying time (DT) variability is explained by this equation. The distribution was independent and identical.

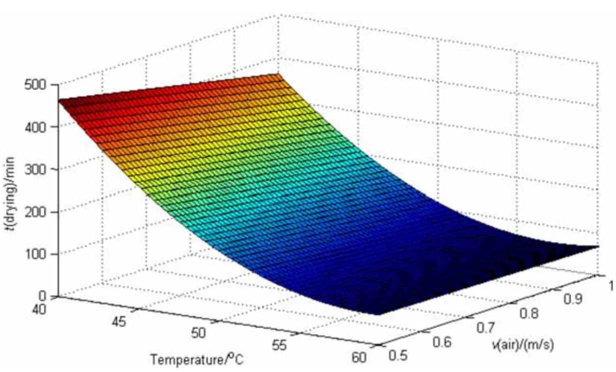

Fig. 5. Drying time of *Echium amoenum* petals as a function of drying temperature and air velocity

Therefore, an optimum combination of drying temperature and air velocity should be established for each product polyphenolic profile to minimize the degradation of these bioactive compounds during the dehydration, leading to the recovery of phenolic compounds and derivatives (*57*). It was concluded from the obtained results that the optimum drying conditions to reach the maximum TPC with the minimum drying time of dried *E. amoenum* petals is the temperature of 60 °C and air velocity of 1.0 m/s.

Conclusions

The effects of different air velocities and temperatures on the drying of *Echium amoenum* petals was evaluated based on the drying parameters such as drying time and moisture ratio. Drying period was completed between 55 and 465 min at different temperatures (40–60 °C) and air velocities (0. 5, 0.75 and 1.0 m/s). Using seventeen different drying models in this study, the statistical parameters including the coefficient of correlation, root mean square error and chi-square values, as well as the constant and coefficients of these models were calculated. Drawing a comparison between the new model and sixteen previous ones, it has been shown that the newly proposed model gives the best results that are mostly in agreement with the experimental data, including the thin-layer drying process. On the other hand, the highest total phenolic content (TPC) was preserved at 60 °C and 1.0 m/s, probably due to the short drying time and therefore, shorter exposure of the phenolics to the thermal effect. The optimum drying conditions of *E. amoenum* petals to extract the maximum TPC are the minimum drying time of 55 min at temperature of 60 °C and air velocity of 1.0 m/s.

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