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Modeling of drying St. John's wort (*Hypericum perforatum* L.) leaves

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Drying of agricultural crops after harvesting is an important operation that helps in preserving product quality and quantity, particularly for medicinal plants and herbs which undergo reduction of essential oils and changes of qualitative properties such as color, both of which influence the economical value of the product. Drying of medicinal plants is a delicate operation for removing product moisture, in order to reduce enzyme activity; thus containing growth of bacteria and pathogens, and preventing product deterioration. Drying process of St. John's wort (*Hypericum perforatum* L.) leaves was studied and modeled in this investigation. Independent variables included temperature at four levels (40, 50, 60 and 70 °C), air velocity at three levels (0.3, 0.7 and 1 m/s), and product depth at three levels (1, 2, and 3 cm). The experiments were performed as factorial with completely randomized design in three replications. Seven drying models, namely Yagcioglu, Page, modified Page, Henderson and Pabis, Lewis, two-term and Verma, were utilized to fit the data. The Page model was found as the best model having the highest R² and lowest χ^2 , RMSE and P-values.

Key words: Drying, St. John's wort, modeling.

INTRODUCTION

The main aims of drying agricultural products are to increase the shelf life, to protect it from biological activities including microbial and enzymatic, and to reduce the weight and volume of the materials in order to facilitate packaging, transporting and storing (Simal et al., 2005). During the drying process, it is important to preserve the texture, color, flavor, and nutritional value of the product. This means reducing it to a safe level of moisture content, to minimize the quantity and quality losses during storage (Hall, 1980). The amount and the type of moisture, have direct effects on drying time. Extensive studies have been conducted on native flora and medicinal herbs in most developed countries (Chenarbon et al., 2010). The research problem regarding medicinal herbs is indentifying and preserving the essential oils especially in different kinds of herbs. One of the most useful medicinal herbs is *Hypericum perforatum* L.

The property of the Iranian species of this crop is high level of Hypericin, as the most important essential oil of its flower and leaves which plays an important role in the medical treatment of various diseases. Its trade level has been \$ 210 m in USA and \$ 570 m throughout the world (Sirvent et al., 2002). This crop has been cultivated in Western Europe. For example in Germany, the production area reached 300 ha in 1997 while it was only 15 ha in 1992 (Buter et al., 1998). Panchariya et al. (2002) examined the drying conditions of black tea at temperatures ranging from 80 to 120 °C and air velocity from 0.25 to 0.65 m/s. Experimental data was evaluated using Lewis, Page, modified Page, Two-term, and

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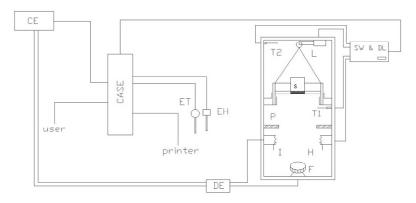


Figure 1. Experimental dryer. (F) fan, (H) heat generator, (S) sample, (T1) Lower thermometer, (T2)Upper thermometer, (Sw) switches, (DL) data logger, (CE) control electronic system, (DE) electronic driver, (EH) environment relative humidity sensor, (ET) environment temperature sensor

Handerson and pabis drying models with nonlinear regression, and the Lewis model was selected as the best model. Arabhosseini et al. (2009) examined drying of Artemisia dracunculus L. leaves at temperatures ranging from 40 to 90 ℃, different relative humidities and an air velocity of 0.6 m/s. Although the diffusion approach equation showed the best fit, but Page model was chosen since it had almost a similar performance but the equation is simpler, as it has only two parameters instead of three in the Diffusion approach model. Dovmaz (2007) assessed the drying behavior of mint leaves at temperatures of 35, 45, 55, and 65 °C and air velocity of 1m/s. He reported that, drying time reduced significantly at higher temperatures. Four drying models were selected to fit the experimental data and the logarithmic model described the drying behavior of mint leaves satisfactorily. Sharma et al. (2007) examined thin layers drying of onions in a dryer using x-ray.

The experiments were conducted at temperatures of 35, 40 and 45 °C and air velocities of 1, 1.25 and 1.5 m/s. The obtained data were fitted to eight drying models. Following statistical analysis, the Page model was proposed as the best model because of highest R^2 and the least χ^2 . In another research, the drying process of mint leaves was evaluated at temperatures levels of 30, 40 and 50 °C and air velocities of 0.5 and 1 m/s and the Page model showed the best fit compared to the other models (Park et al., 2002). The present study was conducted to determine the best model for drying of *H. perforatum* L. leaves which has not been reported before. This information is helpful in designing an effective drying process to preserve quality properties and effective chemical compounds.

MATERIALS AND METHODS

Drying equipment

Three Kiln type laboratory dryers were utilized to conduct the drying

experiments. Drying chamber is a $40 \times 40 \times 50$ cm container located 70 cm above the heating elements (Figure 1). Each dryer has two electric elements to generate the required heat, one of them controlled by a digital thermostat and the other controlled manually. Hot air flow is produced by a blower located under the elements, providing an adjustable flow rate in the range of 180 to 220 m³/h using a dimmer. Two sensors are mounted in the upper and lower parts of the dryer, to measure the temperature of the drying air before and after the samples location.

Prior to starting of each experiment, air temperature was adjusted by the thermostat and the dryers were activated to reach the required temperature. Data collection for thin layer drying experiments was performed through weighing of samples at 5 min intervals using a ±0.00l g digital balance (Sartorius, model PT210, Germany) The mean value of the sample dry weight was used for calculations. Weighing of the samples continued until three consecutive readings showed the same value. Initial and final sample moisture contents were determined gravimetrically before and after the drying experiment. Moisture content of the samples was determined by drying in a vacuum dryer (model Galen Kamp) at 70 °C, 150 mbar, for 8 h (Tsami et al., 1990). The velocity of drying air was adjusted to the desired level by adjusting the blower motor and measured by an anemometer (AM- 4201, Lutron) with an accuracy of ±0.1 m/s. During the experiments the ambient air temperature and RH variations in the laboratory were measured to be between 25 to 29°C and 31 to 33%, respectively. Given the small size of the samples, 35 by 35 cm metal micro-pore meshes were used as tray to keep the samples in the dryers. Aluminum frames of 35 by 35 cm cross section with 1, 2 and 3 cm height were placed on the porous plates to obtain the desired bed depths. The samples were placed in the frame while a metal mesh was used on the frame to avoid any sample skip by air current flowing.

Drying process

Samples of St. John's wort were obtained from the Medicinal Herb Research collection of Jahad-e-Daneshgahi in May and June, 2009. The plants were harvested just before flowering and the leaves were immediately removed from the stems. The leaves were cut and stored separately in plastic bags and refrigerated at temperature of 4 ± 1 °C to prevent microbial activity. Moisture content of the leaves was found to be 61% db. In this study, the independent variables were drying temperature at 40, 50, 60, and 70 °C, air velocity at 0.3, 0.7, and 1 m/s and bed depth at 1, 2, and 3 cm levels. Thus, there were 36 treatments replicated three times.

Model name	Model equation	References		
Lewis	MR = exp(-kt)	(Lewis, 1921)		
Henderson and Pabis	MR = a exp(-kt)	(Westerman et al.,1973)		
Page	$MR = exp(-kt^n)$	(Page, 1949)		
Modified Page	$MR = exp(-(kt)^{n})$	(White et al., 1981)		
Yagcioglu	$MR = a \exp(-kt) + c$	(Yagcioglu et al., 1999)		
Verma	$MR = a \exp(-kt) + (1-a) \exp(-gt)$	(Verma et al., 1985)		
Two-term	$MR = a \exp(-k_0 t) + b \exp(-k_1 t)$	(Gunhan et al., 2004)		

Table 1. Mathematical equations used for modeling of the drying process.

Table 2. Equations of indicators for evaluation of the dryingmodels (San Martin et al., 2001).

Indicator	Equation
P- value	$P = \frac{100}{N} \sum \frac{ M_i - M_{pre} }{M_i}$
<i>x</i> ²	$x^{2} = \frac{\sum_{t=1}^{n} \left(MR_{exp} - MR_{pre}\right)^{2}}{N-n}$
RMSE	$RMSE = \sqrt{\frac{\sum_{i=1}^{n} (MR_{pre} - MR_{exp})^2}{N}}$

Dependent variable was drying time needed to determine the proper model for thin layer drying of St. John's wort leaves. A factorial experiment design was laid out in completely randomized design with three replications. All data were subjected to analysis of variance and Duncan's multiple range test was used to compare the treatment means.

Mathematical modeling of drying

For modeling of thin layer drying of St. John's wort, the moisture ratio was calculated using Equation 1.

$$M\bar{R} = \frac{M_{\rm f} - M_{\rm f}}{M_0 - M_{\rm g}} \tag{1}$$

In which MR is moisture ratio (dimensionless), Mt is moisture content at time t (d.b%), Mo is initial moisture content (d.b%), and Me is equilibrium moisture content (d.b%). For determining the final moisture content, the samples were placed in a vacuum oven at 70 °C and 150 mbar for 8 h. The samples were weighted before and after drying and their moisture content was determined using Equation 2.

$$M_{\sigma} = \frac{W_{iv} - W_{il}}{W_d}$$
(2)

In which Mc is moisture content (d.b%), Ww is weight of sample (kg) and Wd is dry matter weight (kg).

The drying models listed in Table 1 were used to examine moisture variation during the drying process and to determine the best model.

In the equations, k, n, a, b, c, g, k0 and k1 are the model coefficients. Non-linear regression method was utilized to fit the data to the selected drying models. For evaluating the goodness of fit, three statistical indicators were used in addition to R² (Table 2). The model having the highest R² and the lowest Root Mean Squares Error (RMSE), χ^2 and P-value was thus determined as the best model. In Table 2 equation parameters are, Mi: moisture content of matter, Mpre: predicted moisture by the model, N: number of observations, n is number of model constants, MRexp: moisture ratio of experimental data, and MRpre: predicted moisture ratio.

RESULTS AND DISCUSSION

In order to determine MR, first the sorption isotherms of *H. perforatum* L. leaves were obtained at various water activities for four temperature levels of 40, 50, 60 and 70 °C. The full range of water activities and temperatures had a significant effect on EMC and with decreasing temperature at a constant relative humidity, EMC increased. Such behavior may be explained by considering the excitation state of molecules. At high temperatures, molecules are in an increased state of excitation, leading to weaker attractive force. This in turn, results in a decrease in the degree of water sorption at a given relative humidity with increasing temperature (Chenarbon et al., 2010). Drying was continued until the equilibrium moisture content was reached. Variation of

Model	RMSE ×10 ⁻¹	χ ² ×10 ⁻²	R ²	P- value (%)	
V = 0.3 m/s					
Lewis	24	11	0.98	9.42	
Henderson and Pabis	29	43	0.98	25.12	
Page	7	10	0.98	8.42	
Modified Page	28	51	0.98	27.39	
Yagcioglu	39	24	0.89	25.55	
Verma	37	36	0.88	19.80	
Two - term	48	19	0.88	20.90	
V = 0.7 m/s					
Lewis	23	11	0.98	8.17	
Henderson and Pabis	42	45	0.98	16.35	
Page	8	11 0.98		5.84	
Modified Page	28	50	0.97	11.84	
Yagcioglu	36	25	0.88	17.94	
Verma	48	29	0.87	14.35	
Two - term	29	18	0.88	17.25	
V = 1.0 m/s					
Lewis	19	10	0.98	7.85	
Henderson and Pabis	49	47	0.98	17.61	
Page	9	10	0.98 0.98	8.25	
Modified Page	54	43		12.20	
Yagcioglu	45	17	0.89	13.17	
Verma	49	16	0.88	26.14	
Two – term	34	17	0.88	17.31	

Table 3. Evaluation of the models at 40 C and air velocities of 0.3, 0.7 and 1 m/s

moisture content with drying time for varying values of the governing parameters (mainly, air temperature, velocity and depth) was then determined. Increasing the velocity of drying air results in decreasing drying times because of increasing convective heat and mass transfer coefficients between the drying air and the product. Increasing the drying air temperature decreased the total drying time since heat transfer increased. At constant drying air velocity and relative humidity, increasing the temperature of the drying air, decreased the total drying time. Effect of temperature is more pronounced than that of the velocity since the diffusion coefficient is a dependent function of air temperature (Ahmet et al., 2009).

All the data series were fitted with eight equations. Tables 3 to 6 show the obtained statistical results of R^2 , RMSE, and p-value for fitting the experimental data to selected drying models in order to determine the best model.

The Verma, Yagcioglu, and Two-term models were eliminated for having R^2 values lower than 0.9 while the Henderson and Pabis and Modified Page models were omitted because of undesirable χ^2 values.

The Lewis model was eliminated due to very high

RMSE. Overall, the Page model showed the best fit having highest R^2 and lowest χ^2 , RMSE, and P-value. Table 7 shows the results of fitting the Page model to data obtained from experimental treatments. The R^2 values are above 0.99 and p-values are below 10% for all temperature and air velocity levels which statistically shows the good fit.

In a similar research, Park et al. (2002) found the Page model as the best model for drying mint leaves because it showed the best fit to experimental data. The study on drying A. dracunculus L. leaves at air temperatures and relative humidities in the range of 40 to70 °C and 11 to 84%, respectively, and constant air velocity of 0.6 m/s showed that Page model came a close second after the Diffusion approach model. The Page equation has only two variables, which makes it similar and thus. more desirable than the other more complicated equations. Therefore the Page equation was selected as the preferred equation for modeling the drying of tarragon (Arabhosseini et al., 2009). Also, in a research about thinlayer drying of onions using x-ray, the Page model was proposed as the best one for having the highest R² and the least χ^2 (Sharma, 2007).

Model	RMSE × 10 ⁻¹	χ ² × 10 ⁻²	R^2	P- value (%)
V = 0.3 m/s				
Lewis	15	10	0.98	5.78
Henderson and Pabis	59	28	0.97	20.43
Page	11	12	0.97	3.32
Modified Page	42	38	0.97	20.41
Yagcioglu	39	19	0.89	13.91
Verma	61	23	0.89	18.12
Two - term	50	24	0.89	12.26
V = 0.7 m/s				
Lewis	16	14	0.98	6.41
Henderson and Pabis	50	42	0.97	10.89
Page	10	13	0.98	3.28
Modified Page	70	27	0.98	13.31
Yagcioglu	83	16	0.88	12.25
Verma	64	14	0.88	16.21
Two - term	60	21	0.87	17.69
V = 1.0 m/s				
Lewis	19	14	0.98	6.24
Henderson and Pabis	54	34	0.97	19.29
Page	8	10	0.98	2.91
Modified Page	48	29	0.98	13.23
Yagcioglu	35	26	0.88	18.41
Verma	47	18	0.88	18.35
Two - term	40	14	0.88	14.49

Table 6. Evaluation of the models at 70 $^{\circ}\mathrm{C}$ and air velocities of 0.3, 0.7 and 1 m/s.

Model	RMSE × 10 ⁻¹	χ ² × 10 ⁻²	R^2	P- value (%)
V = 0.3 m/s				
Lewis	14	10	0.98	7.25
Henderson and Pabis	46	25	0.98	25.26
Page	8	9	0.97	2.49
Modified Page	31	27	0.98	23.50
Yagcioglu	45	17	0.89	20.67
Verma	39	19	0.88	15.34
Two - term	24	13	0.88	17.41
V = 0.7 m/s				
Lewis	20	17	0.98	3.92
Henderson and Pabis	42	37	0.98	11.86
Page	7	18	0.98	3.91
Modified Page	45	36	0.98	19.27
Yagcioglu	37	16	0.89	12.35
Verma	39	18	0.88	11.99
Two - term	51	17	0.88	14.08
V =1.0 m/s				

Lewis	15	8	0.98	6.23
Henderson and Pabis	26	21	0.97	10.24
Page	6	8	0.98	2.15
Modified. Page	30	28	0.97	11.89
Yagcioglu	27	18	0.89	16.39
Verma	19	19	0.89	25.49
Two - term	20	18	0.89	23.58

Table 6. Evaluation of the models at 70 °C and air velocities of 0.3, 0.7 and 1 m/s (continued).

Table 7. Coefficient of the Page equation fitted to drying data.

Temperature (℃)	Velocity (m/s)	k	n	RMSE ×10 ⁻¹	P- value (%)	X ² × 10 ⁻²	\mathbf{R}^2
	0.3	0.021	0.783	10.70	5.56	30.80	0.997
40	0.7	0.014	0.873	19.20	3.45	78.14	0.999
40	1	0.008	0.901	21.84	4.23	28	0.998
	0.3	0.016	0.768	36.41	4.1	28.23	0.998
	0.7	0.022	0.812	58.61	4.26	52.70	0.998
50	1	0.038	0.879	9.9	4.30	61.95	0.999
	0.3	0.040	0.793	20.41	6.84	46.65	0.997
60	0.7	0.039	0.801	30.71	5.92	29.85	0.997
	1	0.36	0.871	45.18	3.21	31.24	0.999
	0.3	0.40	0.867	26.35	3.37	14.36	0.998
70	0.7	0.042	0.845	17.25	2.64	17.25	0.998
	1	0.037	0.978	5.18	2.20	13.12	0.999

Conclusion

Among seven drying models, the Page equation showed the best fit for drying of St. John's wort leaves. Thus, this model is suitable as a relevant equation for drying of *H. perforatum* L. The n and k parameters were estimated as functions of temperature for St. John's wort leaves. This model is suitable to estimate the moisture content during drying, in order to determine drying time and energy consumption. It is also applicable for designing of relevant dryer for this type of medicinal plant.

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