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Thin Layer Drying of Bay Leaves (*Laurus nobilis* L.) in Conventional and Microwave Oven

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ABSTRACT

The thin layer oven drying behaviour of bay leaves at temperatures of 50, 60 and 70°C in conventional built-in oven and 180W power level in microwave oven was investigated. Eight different thin layer drying models namely Lewis, Henderson and Pabis, Page, two-term, two-term exponential, parabolic, logarithmic and Midilli *et al.* were fitted to experimental drying data. The highest adjusted R-square with the lowest reduced chi-square and root mean square error were selected as statistical criteria to evaluate how well the tested models fit the drying data. Midilli *et al.* model was considered to be satisfactory to represent the thin layer oven drying of bay leaves. Effective diffusion coefficient (D_{eff}) was found between 1.52×10^{-9}-8.08×10^{-9} m^2/s for conventional oven. The temperature dependent activation energy (E_a) was determined as 40.10 kJ/mol for conventional oven.

Key Words: Drying, Bay leaves, Thin layer, Modeling, Effective diffusion coefficient

Defne yaprağının (*Laurus nobilis* L.) Konvansiyonel ve Mikrodalga Fırında İnce Tabaka Kurutulması

ÖZET

Defne yaprağının konvansiyonel fırında 50, 60 ve 70°C'de ve mikrodalga fırında 180W güç seviyesinde ince tabaka kuruma davranışı incelenmiştir. Lewis, Henderson ve Pabis, Page, two-term, two-term exponential, parabolic, logarithmic ve Midilli *et al.* olarak literatürde tanımlanan sekiz farklı ince tabaka kuruma modeli deneysel verilere uygulanmıştır. En yüksek düzeltilmiş belirleme katsayısı ile en düşük indirgenmiş ki-kare ve en düşük kök ortalama kare hatası deneysel verilerin hangi modele daha uygun olduğunu belirleme ölçütü olarak seçilmiştir. Midilli *et al.* modeli defne yaprağının fırında kurutulmasını temsil edecek düzeyde yeterli bulunmuştur. Konvansiyonel fırın için etkin difüzyon katsayısı (D_{eff}) değerleri 1.52×10^{-9}-8.08×10^{-9} m^2/s arasında bulunmuştur. Ayrıca sıcaklığa bağımlı aktivasyon enerjisi konvansiyonel fırın için 40.10 kJ/mol olarak bulunmuştur.

Anahtar Kelimeler: Kurutma, Defne yaprağı, İnce tabaka, Modelleme, Etkin difüzyon katsayısı

INTRODUCTION

The bay leaf (*Laurus nobilis* L.), also known as laurel leaf, is an evergreen perennial tree leaf and indigenous to many Mediterranean and European countries. Fresh or dried leaves are often used for flavouring in various dishes and pickles with its strong aroma. In traditional medicine, bay leaves have been used to treat bronchitis, dermatological disorders, inappetency, and alleviation of rheumatism pain. As an alternative pharmaceutical, bay leaves were effective in reducing blood glucose and total cholesterol in people with type-2 diabetes [1, 2], and improvement and prevention of insulin resistance [3]. Chloroform fraction of these leaves is a potential drug candidate by protection of cerebral ischemia neuronal damage [4]. Bioactive compounds of bay leaves derived from essential oil or extracts has high total antioxidant activity and high free radical scavenging activity [5], as well as strong antibacterial effect against all tested food borne spoilage and pathogenic bacteria [6, 7, 8].

Suitable dryer selection is very important for protecting against the loss of volatile compounds in bay leaves. Increasing drying temperature may result in a decrease of most volatiles [9], and essential oil content [10]. According to Diaz-Maroto *et al.* [11], oven drying at 45°C and air drying at ambient temperature caused the minimum loss in volatiles. In recent literature there have been many studies about drying of herbs and spices rich in bioactive compounds as well as modeling of drying to evaluate and predict the process parameters [12, 13, 14, 15, 16]. However there is limited information in modeling of conventional oven drying and microwave drying and there is no sufficient information about the comparison of these drying methods by thin layer modeling at the same time.

With this study, drying mechanism of bay leaves using conventional and microwave oven was determined, besides eight different thin layer models were tested to find the best fitting model in order to simulate the drying process. Total colour difference, rehydration ratio and water activity was also determined to evaluate the main quality parameters of dried bay leaves.

MATERIALS and METHODS

Bay leaves (*Laurus nobilis* L.) having an average thickness of 0.3 mm was picked up from Ege University immediately before experiment. Leaves without defect were washed and excess water was removed with tissue paper prior to drying. AOAC 934.01 vacuum oven method was used to determine the initial moisture contents of bay leaves [17]. The initial moisture content of fresh bay leaves was 52.06±1.27% (wet basis).

Digital built-in conventional oven model no. NE66209D0 (Vestel, Turkey) and microwave oven model no. MD595 (Arcelik, Turkey) were used in drying experiments. Conventional oven was preheated until the set temperatures namely 50, 60 and 70°C have been reached. Lower and upper heating element function (without fan) was selected and bay leaves were uniformly placed into the aluminium oven tray (32x34 cm) in the middle rack position. During preliminary trials, rapid browning and shrinkage occurred at higher power levels in microwave oven, so microwave drying was performed only at 180W power (minimum level). The samples were weighed every 10 minutes for conventional oven treatment and every 2 minutes for microwave oven treatment until constant weight was observed. All tests were performed in triplicate.

Some quality parameters of bay leaves were tested to identify the differences between samples. Water activities of dried leaves were measured using Testo AG 400 (Germany) water activity measurement device. Dried leaves were grinded prior to measurement in a laboratory type blender (Waring Inc., USA). The colour of fresh and dried leaves according to CIE colour space (L*, a*, b*) was measured using a Minolta Chroma Meter CR-400 (Konica Minolta Sensing Inc., Japan). The effect of drying condition on colour was calculated from the total colour difference (ΔE*) according to the given equation;

$$\Delta E^* = \sqrt{(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2} \qquad (1)$$

Statistical analysis was performed using SPSS software version 16.0 (SPSS Inc., USA). One-way analysis of variance was used to compare the mean values and Duncan post hoc multiple comparison test was applied with a significance level of $p<0.05$ to evaluate the differences between samples.

Rehydration analyses were performed according to the method of Doymaz [18] with slight modifications. About 2 g of dried product was placed into the beaker containing 1/100 distilled water (w/w) at room temperature. Samples were taken out after 4 h and excess water was removed using tissue paper. The rehydration ratio was calculated according to following equation;

$$\text{Rehydration ratio} = \frac{\text{total mass after rehydration}}{\text{total mass before rehydration}} \qquad (2)$$

Mathematical Modeling of Drying Data

Fick's second law of diffusion is generally used to describe moisture diffusion in a solid particle;

$$\frac{\partial M}{\partial t} = D_{eff}\frac{\partial^2 M}{\partial x^2} \qquad (3)$$

where M is local moisture content on dry basis, D_{eff} is effective diffusion coefficient, t is time and x is spatial coordinate [19]. The diffusion equation for the falling-rate drying period for a slab can be derived assuming that the initial moisture distribution is uniform, shrinkage is negligible and moisture is migrating only by diffusion. Solution of this equation for an infinite slab can be calculated according to the following formula;

$$MR = \frac{M - M_e}{M_0 - M_e} = \frac{8}{\pi^2}\sum_{n=0}^{\infty}\frac{1}{(2n+1)^2}\exp\left[(2n+1)^2\frac{\pi^2}{4}\frac{D_{eff}t}{L^2}\right] \qquad (4)$$

where MR is the moisture ratio, M_0 is initial moisture content (kg water/kg dry solid) at $t=0$, M_e is equilibrium moisture content (kg water kg^{-1}dry solid), M is the moisture content at time t (kg water/kg dry solid) and L is the thickness of the slab (m) for the solids when evaporation occurs from only one face [20]. At sufficiently large drying times, only the leading term in the series of expansion is taken into account to calculate the effective diffusion coefficient;

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

Eight different thin layer drying models which are often used in literature were applied to drying data (Table 1). These expressions were tested in MATLAB software version 7.7.0 (MathWorks Inc., USA) using curve-fitting tool box. To evaluate the goodness of fit, adjusted R-square (*adj-R^2*), reduced chi-square (χ^2) and root mean square error (*RMSE*) were calculated with the following equations;

$$adj-R^2 = 1-(1-R^2)\frac{N-1}{N-m-1}, \qquad R^2 = \frac{\sum_{i=1}^{N}(MR_i - MR_{pre,i}).\sum_{i=1}^{N}(MR_i - MR_{exp,i})}{\sqrt{\left[\sum_{i=1}^{N}(MR_i - MR_{pre,i})^2\right].\left[\sum_{i=1}^{N}(MR_i - MR_{exp,i})^2\right]}} \tag{6}$$

$$\chi^2 = \frac{\sum_{i=1}^{n}(MR_{exp,i} - MR_{pre,i})^2}{N-n} \tag{7}$$

$$RMSE = \left[\frac{1}{N}\sum_{i=1}^{N}[(MR_{exp,i} - MR_{pre,i})^2]\right]^{\frac{1}{2}} \tag{8}$$

where $MR_{pre,i}$ expresses the predicted moisture ratio, $MR_{exp,i}$ expresses the experimental moisture ratio, N is the number of observations, m is number of regression parameters, and n is the number of constants. The model having the highest *adj-R^2*, and lowest χ^2 and *RMSE* was chosen as the best fitting model.

Table 1. Mathematical models fitted to drying curves

Model No	Model Name	Model Expression	Reference
1	Lewis	$MR = \exp(-kt)$	[21]
2	Henderson and Pabis	$MR = a\exp(-kt)$	[22]
3	Page	$MR = \exp(-kt^n)$	[23]
4	Two term	$MR = a\exp(-k_0 t) + b\exp(-k_1 t)$	[24]
5	Two term exponential	$MR = a\exp(-kt) + (1-a)\exp(-kat)$	[25]
6	Parabolic	$MR = a + bt + ct^2$	[26]
7	Logarithmic	$MR = a\exp(-kt) + c$	[27]
8	Midilli *et al.*	$MR = a\exp(-kt^n) + bt$	[28]

Calculation of Effective Diffusion Coefficient and Activation Energy

The effective diffusion coefficient was calculated from Equation (9), the slope *m* was determined from the plot of *ln*(*MR*) versus time;

$$m - \frac{\pi^2 D_{eff}}{4L^2} \tag{9}$$

Temperature dependence of the effective diffusion coefficient (D_{eff}) may described by Arrhenius equation as follows;

$$D_{eff} = D_0 \exp\left(-\frac{E_a}{RT}\right) \tag{10}$$

where E_a is the activation energy (kJ/mol), T is the absolute temperature (K), D_0 is a reference diffusion coefficient (m^2/s) and R is the universal gas constant (kJ/molK) [29]. The activation energy can be determined from the plot of *ln* (D_{eff}) versus *1/T*.

RESULTS and DISCUSSION

Bay leaves having an average 52.06±1.27% (wet basis) initial moisture content were dried at 50, 60, and 70°C in conventional oven using lower and upper heating element function where the effect of air velocity is neglected and at 180W in microwave oven until the constant weight -which is assumed to be the equilibrium moisture content- have been reached. The drying curves which were obtained from the average of MR's are presented in Figure 1. Only the falling-rate period was observed for these drying conditions. Gunhan *et al.* [30] obtained the similar behaviour for bay leaves that were dried in a hot air dryer. As can be seen from Figure 1, drying time decreases with increasing temperature. Compared to conventional oven drying, substantial decrease in drying time (nearly 4 times lower) was observed for microwave drying. The major part of the moisture was reduced in the early stages of drying and it gradually decreased in later stages.

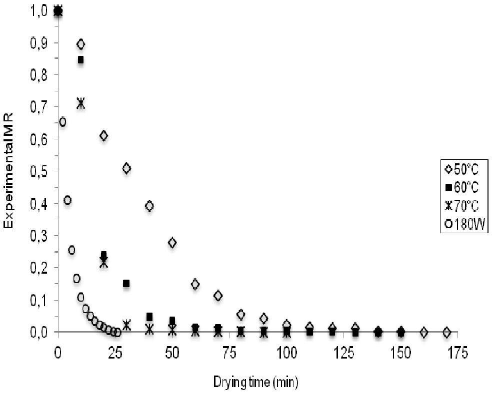

Figure 1. Drying curves at given conditions

The results of water activity are presented in Figure 2. Increasing values of drying temperature decreased the water activities. These values are in safe limits that can retard or eliminate enzyme activity, mold and bacteria growth or browning reactions.

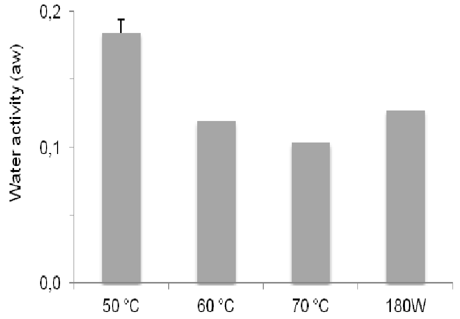

Figure 2. Water activity values at given drying conditions

The averages of total colour difference and L*, a* and b* values are shown in Table 2. There are several factors influencing the quality parameters of dried product. Some chemical reactions such as browning reactions and lipid oxidation might alter the final colour [19]. Although there is no significant difference in total colour difference, the greenness (-a* values) of the dried samples was increased with decreasing the drying time or increasing the drying temperature. Microwave dried bay leaves due to its shortest drying time (26 min.) had the highest greenness value.

Table 2. Colour values of dried bay leaves

Drying condition	*L**	*a**	*b**	*ΔE**
50°C	44.93^{b}	-5.88^{d}	14.56^{b}	12.17^{a}
60°C	49.87^{c}	-7.18^{c}	12.41^{a}	16.06^{b}
70°C	43.82^{a}	-8.19^{b}	16.32^{c}	12.03^{a}
180W	$44.12^{a,b}$	-11.26^{a}	$16.06^{b,c}$	12.55^{a}

^{a,d} Values followed by different letters in the same column are significantly different ($p<0.05$).

Rehydration can be considered as a measure of the cellular and structural changes to the material caused by dehydration, and preceding treatments [31]. Processing conditions, sample preparation, sample composition can also influence this process [19]. The rehydration ratios are shown in Figure 3. It demonstrates that increasing drying temperature can also increase the rehydration ratios of dried bay leaves. These results are in agreement with the study of Doymaz [18]. However there is still some research needed to confirm this theory.

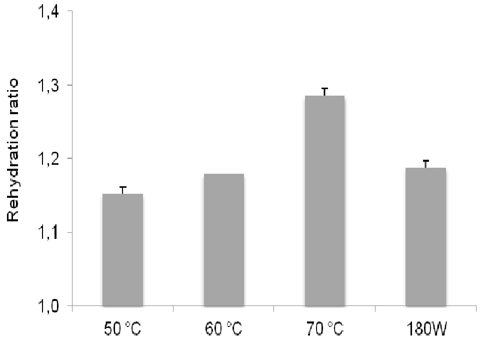

Figure 3. Rehydration ratios at given drying conditions

Mathematical Modeling of Drying Data

Statistical results of tested thin layer drying models are presented in Tables 3-6. It can be seen that there is a good agreement between experimental data and tested mathematical models. The best fitting model was selected according to the highest adjusted R-square and reduced chi-square and root mean square error calculated from equations (6), (7) and (8). With respect to given selection criteria, Midilli *et al.* model for 50°C, two-term exponential model for 60°C, Midilli *et al.* model for 70°C, and Lewis model for 180W drying gave the best fit. However, Midilli *et al.* model was chosen for the representation of thin layer drying of bay leaves with its comparably high $adj\text{-}R^2$ (>0.98) and low χ^2 (<0.002) and *RMSE* (<0.05) for whole drying conditions to compare the results easily. Midilli *et al.* model was also obtained by other researches for dill and parsley [32], mint [12], saffron stigmas [33]. Comparison of experimental and Midilli *et al.* model predicted moisture ratios are presented in Figure 4. This figure shows that there is a good correlation between the experimental and predicted values.

Table 3. Statistical results and model parameters of drying at 50°C

Model no	Model constants	$Adj\text{-}R^2$	χ^2	*RMSE*
1	k=0.02672	0.9773	0.00236	0.04859
2	a=1.079, k=0.02854	0.9818	0.00190	0.04356
3	k=0.006656, n=1.363	0.9954	0.00048	0.02337
4	a= -3.166, k_0=0.01596, b=4.224, k_1=0.01829	0.9876	0.00129	0.03585
5	a=1.930, k=0.0395	0.9947	0.00056	0.02357
6	a=0.9598, b=-0.0161, c= 6.421x10^{-5}	0.9734	0.00277	0.05260
7	a= 1.105, k=0.02545, c= -0.0422	0.9859	0.00146	0.03826
8	a=1.005, k=0.007103, n=1.347, b=-1.855x10^{-5}	0.9957	0.00048	0.02196

Table 4. Statistical results and model parameters of drying at 60°C

Model No	Model Constants	*Adj-* R^2	χ^2	*RMSE*
1	k=0.05371	0.9392	0.00588	0.07667
2	a=1.081, k=0.05702	0.9407	0.00574	0.07578
3	k=0.00102, n=2.353	0.9840	0.00155	0.03939
4	a= -1.451, k_0=2.915, b=2.451, k_1=0.1075	0.9948	0.00050	0.02246
5	a=2.878, k=0.1155	0.9949	0.00050	0.02232
6	a=0.8087, b=-0.01842, c=9.289x10^{-5}	0.7808	0.02121	0.09180
7	a= 1.093, k=0.05504, c= -0.01386	0.9375	0.00605	0.07777
8	a=1.022, k=0.001286, n=2.285, b=7.207x10^{-5}	0.9823	0.00171	0.04138

Table 5. Statistical results and model parameters of drying at 70°C

Model No	Model Constants	*Adj-* R^2	χ^2	*RMSE*
1	k=0.06583	0.9506	0.00593	0.07700
2	a=1.058, k=0.06853	0.9487	0.00616	0.07851
3	k=0.002241, n=2.178	0.9999	0.00002	0.00401
4	a=10.08, k_0=0.04104, b=-9.05, k_1= 0.03884	0.9472	0.00635	0.07966
5	a=2.842, k=0.1321	0.9989	0.00014	0.01175
6	a=0.8957, b= -0.02959, c=2.182x10^{-4}	0.8664	0.01604	0.12670
7	a= 1.084, k=0.06386, c= -0.02878	0.9468	0.00639	0.07992
8	a=0.9999, k=0.002225, n=2.181, b=2.993x10^{-5}	0.9999	0.00002	0.00399

Table 6. Statistical results and model parameters of drying at 180W

Model no	Model constants	*Adj-* R^2	χ^2	*RMSE*
1	k=0.2212	0.9997	0.00003	0.00509
2	a=1.003, k=0.222	0.9997	0.00003	0.00518
3	k=0.2171, n=1.011	0.9997	0.00003	0.00512
4	a= 0.2769, k_0=0.2075, b=0.7272, k_1=0.2282	0.9996	0.00003	0.00567
5	a=0.8648, k=0.2258	0.9997	0.00003	0.00529
6	a=0.8451, b= -0.09587, c=0.002569	0.9233	0.00680	0.08244
7	a= 1.003, k=0.2228, c= 0.00111	0.9997	0.00003	0.00536
8	a=1.002, k=0.2167, n=1.015, b=9.266x10^{-5}	0.9997	0.00003	0.00541

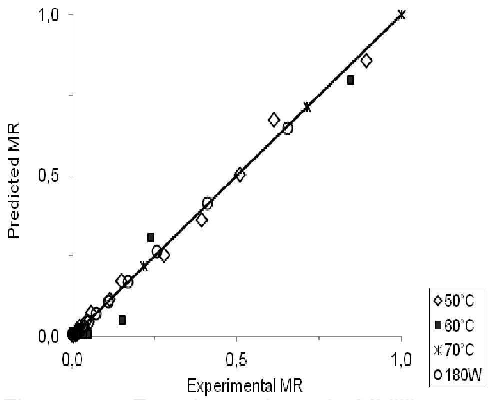

Figure 4. Experimental and *Midilli et al.* predicted moisture ratios

Calculation of Effective Diffusion Coefficient and Activation Energy

The effective diffusion coefficients were calculated from Equation (9) with a high coefficient of determination (R^2>0.98), and the results are shown in Table 7. Erbay and Icier [34] reported that D_{eff} of foods that were dried in a convective type dryer, generally fall within the region of 10^{-10} to 10^{-8} m^2/s (86.2%). The present findings seem to be consistent with those mentioned literature.

Table 7. Effective diffusion coefficients at given conditions

Temperature/Power	D_{eff} (m^2/s)
50°C	1.52x10^{-9}
60°C	2.11x10^{-9}
70°C	3.64x10^{-9}
180W	8.08x10^{-9}

Temperature dependence of effective diffusion coefficient was expressed by Arrhenius equation and the relation between ln (D_{eff}) and $1/T$ was linear (R^2>0.97). The activation energy was calculated as 40.10 kJ/mol for conventional oven. This value is similar to those obtained by other researchers such as; 35.05 kJ/mol for dill leaves [32], 43.92 kJ/mol for parsley leaves [32], and 38.78 kJ/mol for cape gooseberries [35]. In the study of Erbay and Icier [34], activation energy of the compiled studies generally accumulates in the range of 18 to 49.5 kJ/mol. In the recent study of Doymaz [36], activation energy of bay leaves dried at similar temperatures in a cabinet dryer was found as 36.48 kJ/mol; however, the effective moisture diffusion coefficient was much more smaller (9.38×10^{-12} to 2.07×10^{-11} m^2/s) than present results.

The thin layer drying kinetics of fresh bay leaves was experimentally determined in conventional and microwave oven. Oven temperature and/or power were selected as variable in this study. Bay leaves reached the equilibrium moisture content between 26-170 minutes depending on the drying process. The best model explaining the drying behaviour of bay leaves was found to be Midilli *et al.* model. In spite of the fact that temperature and moisture diffusion during microwave drying is a much more complex process, all the tested models were sufficient to explain the experimental moisture ratio change with respect to time. The aforementioned quality parameters which are water activity, colour -especially greenness- and rehydration ratios were significantly changing with the increasing drying temperature (p<0.05).

Nomenclature	
a, b, c, k, k_0, k_1, n	Drying constants
MR	Moisture ratio (dimensionless)
M	Moisture content at time t (kg water/kg dry solid)
M_e	Equilibrium moisture content (kg water/kg dry solid)
M_0	Initial moisture content (kg water/kg dry solid)
T	Drying time (s)
M	Slope
L	Thickness of material (m)
E_a	Activation energy (kJ/mol)
R	Universal gas constant (8.314 kJ/molK)
T	Absolute temperature (K)
$adj\text{-}R^2$	Adjusted R-square
χ^2	Reduced chi-square
$RMSE$	Root mean square error
N	Number of observations
m	Number of regression parameters excluding intercept
n	Number of constants
Subscripts	
exp	Experimental
pre	Predicted
eff	Effective

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