ESTIMATION OF TEA LEAVES MASS TRANSFER PARAMETERS UNDERGOING MICROWAVE HEAT TREATMENT

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***Abstract*— In the present study, tea leaves (Camellia sinensis) were dried in the domestic microwave oven in order to determine the effect of microwave output power and sample amount on moisture ratio, drying time and effective moisture diffusivity. Five different microwave output power (180-900 W) and five different sample amount (20-100 g) values were used in the drying experiments. The drying data were fitted to some empirical and semi empirical models in order to determine the kinetic parameters. Among of the models proposed, the Page model gave a better fit for all drying conditions applied. The effective moisture diffusivity values were calculated by using the method of slopes at various microwave output powers and sample amounts. Moreover, the activation energy was calculated using an exponential expression based on Arrhenius equation.**

***Keywords*— Microwave drying; Tea leaves; Drying kinetics; Activation energy; Effective moisture diffusivity**

I. INTRODUCTION

The demand for high-quality dried food products is permanently increasing all over the world. Drying, in general, means of removal of water from the material. In order to allow longer periods of storage, minimize packaging requirements, reduce shipping weights and preserve seasonal plants and make them available to consumers during the whole year food products are dried. On the other hand, some important structural and physicochemical changes which affect the final product quality are occurred during dehydration. Microwave drying is an alternative method due to the precise process control, fast startup and shutdown conditions; and it also reduces the drying time and prevents food from decomposing (Barbosa-Canovas and Vega-Mercado, 1996; Decareau, 1985; Decareau, 1992; Zhang *et al*., 2006).

Tea is one of the most extensively consumed beverages because of its health-promoting benefits in terms of its antioxidant properties. Drying plays as a vital part of tea processing because it affects its antioxidant content and appearance that determines the commercial value of the tea (Chong and Lim, 2012).

The study of the drying kinetic of foods during microwave drying has recently been a subject of interest for various investigators. Some of the previous studies on microwave drying kinetics were reported for basil (Demirhan and Ozbek, 2010a), celery leaves (Demirhan and Ozbek, 2011; Alibas, 2014a), corn (Nair *et al*., 2011), ginger (Hussain *et al*., 2010), mallow (Alibas and Koksal, 2014), mint (Ozbek and Dadali, 2007), mushroom (Lombrana *et al*., 2010), nettle (Alibas, 2010), olive pomace (Sadi and Meziane, 2015), parsley (Soysal and Oztekin, 2006), pumpkin (Wang *et al*., 2007a; Alibas, 2007), purslane (Demirhan and Ozbek, 2010b), rosehips (Evin, 2011), quince (Baltacioglu *et al*., 2015), spinach (Dadali *et al*., 2007a), thyme (Sarimeseli *et al*., 2014) and tomato slices (Celen and Kahveci, 2013).

In recent years, microwave drying has gained popularity as an alternative drying method in the food industry. However, there have been no reports on microwave drying characteristics of Turkish tea. The microwave heating could significantly enhance the sensory quality of tea. The reduction in time and energy consumption by using microwave in tea drying could make a significant contribution to tea industry. Also, tea is one of the most extensively consumed beverages because of its health promoting benefits in terms of its antioxidant properties. Drying plays as vital part of tea processing because it affects its antioxidant content and appearance that determines the commercial values of tea. Thus, it is necessary to study the microwave heat treatment on the drying kinetics of tea. The aim of the present study was to investigate the effect of microwave output power and sample amount on the drying kinetics of tea leaves, to compare the experimental data obtained during the drying process with the predicted values obtained by using various drying models, to calculate the effective moisture diffusivity, to calculate the activation energy and to derive a relationship between the drying rate constant and the effective moisture diffusivity.

II. METHODS

A. Materials

Plants of fresh tea samples were collected from tea garden in Artvin in Turkey. Fresh tea samples firstly were washed; then to equilibrate the moisture they were stored in a 4±0.5°C refrigerator for about one day. 50 g samples were dried in an oven (Memmert UM-400) to determine the initial moisture content. This process was repeated four times and the initial moisture content of tea leaves was calculated as 3.30 kg water/kg d.b as an average of the results obtained. On the other hand, for calculation of the effective moisture diffusivity values, the thickness of the tea leaves was measured by a micrometer (Leica ICM 1000, Germany) (30 samples of leaves was measured) and the average thickness of tea leaves found as 0.37 (±0.04) mm.

B. Drying Equipment and Drying Procedure

Drying treatment was performed in a domestic digital microwave oven (Arcelik MD 594, Turkey). The microwave oven has the capability of operating at five different microwave output powers, being 180, 360, 540, 720 and 900 W. The adjustment of microwave output power and processing time was done with the aid of a digital control facility located on the microwave oven.

During drying experiments, each sample was put on the rotating glass at the center of the microwave oven and moisture loss was periodically measured by weighing on the digital balance. Each weighing process was completed in less than 10 s during the drying process. The microwave power was applied until the weight of the sample reduced to a level corresponding to moisture content of about 0.1 kg water/kg d.b. Three replications of each experiment were performed at same operating conditions, and the data given was an average of these results. The reproducibility of the experiments was within the range of ±5%.

C. Mathematical Modelling of Drying Data

In order to determine the moisture ratio as a function of drying time, eight different thin-layer drying models (Page's, Henderson & Pabis, Lewis, Two term, Midilli et al., Wang & Sing, Logarithmic, Modified Page) were selected. The moisture ratio of tea leaves were calculated by using the following equations:

$$MR = \frac{M_t - M_e}{M_o - M_e} \tag{1}$$

In Eq. 1, the equilibrium moisture content (M_e) was assumed to be zero for microwave drying (Dadali *et al.*, 2007b; Wang *et al.,* 2007b; Maskan, 2010).

D. Effective Moisture Diffusivity

The effective moisture diffusivity of a food material characterizes the intrinsic mass transfer property of moisture and explained with Fick's diffusion equation. In drying process, it can be assumed that diffusivity is the only physical mechanism to transfer the water to surface (Wang *et al.,* 2007b; Dadali *et al*., 2007c). Effective moisture diffusivity of the material affected from composition, moisture content, temperature and porosity of the material and it is used due to the limited information on the mechanism of moisture movement during drying and complexity of the process (Abe and Afzal, 1997). The tea leaves were assumed as a slab for the solution of Fick's diffusion equation. The effective moisture diffusivity was therefore calculated by the following straight-line equation (Dadali *et al.,* 2007a; Sobukola *et al.,* 2007; Crank, 1975)

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

The effective moisture diffusivities are typically determined by plotting experimental drying data in terms of ln(*MR*) versus time.

E. Statistical Analysis

For the evaluation of the statistical data, the software package Curve Expert 1.4 was used. The parameters were evaluated by the nonlinear least squares method of Marquardt-Levenberg until minimal error was achieved between experimental and calculated values.

The coefficient of the determination (R^2) is primary parameter for determining the most suitable equation. The standard error of estimated (σ) is defined as the parameter represents the average distance that the observed values fall from the regression of the model and is given by;

$$\sigma^2 \approx s^2 = \frac{\sum_{m=1}^{N_d} (C_m^{obs} - C_m^{cal})^2}{\left(N_d - p\right)} \tag{3}$$

where; N_d is total number of observations, p is the number of parameters and s^2 is the variance. The ideal value of the standard error of estimated is zero.

III. RESULTS AND DISCUSSIONS

A. Effect of microwave output power on the drying kinetics of tea leaves

The effect of microwave output power were investigated by drying 20 g of tea leaves at different microwave output powers ranging from 180 W to 900 W. The moisture ratio values of tea leaves versus drying time were shown in Fig. 1. As the microwave output power was increased, the drying time of samples was significantly decreased, as expected. In the microwave drying process, the drying time was shortened by 77% by working at 900 W instead of 180 W. The experimental results showed that there is no constant rate period observed during microwave drying of tea leaves and drying takes place only in the falling period in which internal liquid diffusion controls throughout. These results were in agreement with the results obtained from other researchers for grape leaves (Alibas, 2014b), coriander (Sarimeseli, 2011), celery leaves (Demirhan and Ozbek, 2011; Alibas, 2014a) and tomato pomace (Al-Harahsheh *et al.,* 2009).

For determination of drying kinetics of tea leaves at various microwave output powers, eight different semi empirical thin-layer drying models presented in Table 1 were used. Among of the models examined, the Page model was observed to be the most appropriate one for all the experimental data with higher value for the coefficient of determination (R^2) and lower standard error (σ) compared with the statistical values obtained for other models. The estimated parameters and statistical analysis of this model for a given drying condition are presented in Table 1. As can be seen from this table, the value of the drying rate constant (*k*) increased with the increase in microwave output power. This implies that with increase in microwave output power the drying curve becomes steeper indicating an increase in drying rate.

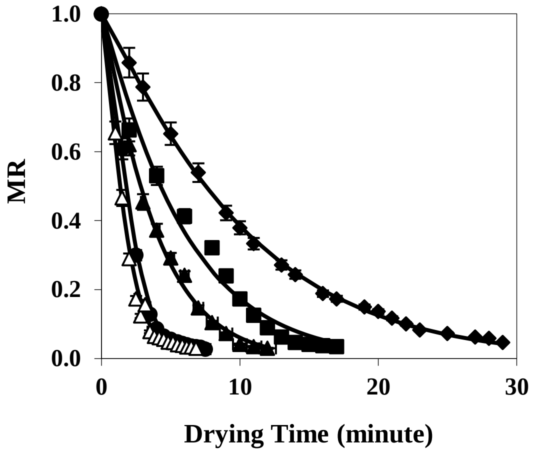

Figure 1: Moisture ratios versus time at various microwave output powers ◆ 180 W, ■ 360 W, ▲ 540 W, ● 720 W, ∆ 900 W, — Kinetic models.

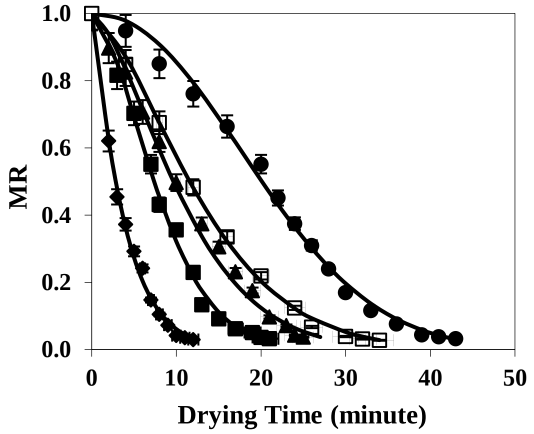

Figure 2. Moisture ratios versus time at various sample amounts ◆ 20 g, ■ 40 g, ▲ 60 g, □ 80 g, ● 100 g, — Kinetic models.

Table 1. The estimated coefficients and statistical analysis of Page model at various microwave output powers

Power (W)	k	n	σ	R^2
180	0.0717	1.1234	0.0077	0.9997
360	0.1432	1.0866	0.0165	0.9985
540	0.1874	1.1099	0.0198	0.9980
720	0.2289	1.4639	0.0227	0.9971
900	0.2627	1.2727	0.0250	0.9958

Table 2. The estimated coefficients and statistical analysis of Page model at various sample amounts

Sample (g)	k	n	σ	R^2
20	0.1874	1.1099	0.0198	0.9980
40	0.1216	1.4084	0.0121	0.9994
60	0.0729	1.5707	0.0252	0.9975
80	0.0439	1.5568	0.0186	0.9988
100	0.0147	1.6051	0.0160	0.9990

B. Effect of sample amount on the drying kinetics of tea leaves

To investigate the effect of sample amount, five sample amounts (20, 40, 60, 80 and 100 g) were dried at a constant microwave output power of 540 W. The values of moisture ratio versus drying time of tea leaves as affected by microwave output power are shown in Fig. 2. As the sample amount was increased, the drying time was increased as well. The microwave drying process which reduced the moisture content of tea leaves to 0.1 g water /g d.b took 12-43 minutes as the sample amount increased from 20 to 100 g, respectively. Similar with the microwave output power effect, the constant drying rate period was again absent and the drying process took place in the falling rate period.

For determination the drying kinetics at different sample amount values, eight different semi empirical thin-layer drying models were used. Among of these models, the Page model was again found the most suitable one for all the experimental data. The estimated parameters and statistical analysis of this model for a given drying condition are presented in Table 2. It was determined that the value of the drying rate constant (k) increased with the decrease in the sample amount.

C. Calculation of the effective moisture diffusivity

The effective moisture diffusivity values calculated by using the method of slopes. The logarithm of moisture ratio values, ln(MR), were plotted against drying time (t) according to the experimental data obtained at various microwave output powers and sample amounts. The effective moisture diffusivity values (D_{eff}) and the statistical parameters of Eq. (2) were presented in Table 3 for various microwave output powers and sample amounts. The effective moisture diffusivity values (D_{eff}), obtained from this study were within the general range of 10^{-11} to 10^{-9} m^2/s for food materials. For comparing the results obtained, no documentary was found in literature that considering the effect of microwave treatment on effective moisture diffusivity for tea leaves.

D. Estimation of Activation Energy

In the present study, the Arrhenius type exponential equation was used in a modified form to illustrate the relationship between the kinetic rate constant and the ratio of the microwave output power to sample amount instead of the temperature for calculation of the activation energy. After evaluation of the data, the dependence of the kinetic rate constant on the ratio of microwave output power to sample amount was represented with an exponential equation Eq. (4) derived by Dadali *et al.* (2007c);

$$k = k_o \exp\left(\frac{-E_a.m}{P}\right) \tag{4}$$

The values of k versus m/P shown in Fig. 3 accurately fit to Eq. (4) with the statistic values of standard error (σ) and coefficient of determination (R^2) of 0.0134 and 0.9879, respectively. Then, k_0 and E_a values were estimated as 0.3073 min^{-1} and 11.54 W/g.

E. Effect of ratio of microwave output power to sample amount on effective moisture diffusivity

It was aimed in this study to predict a relationship between the effective moisture diffusivity and the ratio of microwave output power to sample amount by following the procedure as mentioned in the previous section. After evaluation of the data, to determine the dependence of the effective moisture diffusivity on the ratio of microwave

Table 3: The estimated effective moisture diffusivity and statistical analysis of linear model at various microwave output powers and sample amounts

Power (W)	Slope	$D_{eff}.10^{10}$ ($m^2.s^{-1}$)	σ	R^2
180	0.0032	0.4163	0.1592	0.9853
360	0.0051	0.6671	0.1580	0.9913
540	0.0067	0.8851	0.1472	0.9929
720	0.0083	1.0938	0.2474	0.9781
900	0.0090	1.1844	0.2450	0.9768
Sample (g)	**Slope**	**$D_{eff}.10^{11}$ ($m^2.s^{-1}$)**	**σ**	**R^2**
20	0.0067	8.8510	0.1472	0.9929
40	0.0041	5.4363	0.1498	0.9933
60	0.0033	4.3070	0.2792	0.9869
80	0.0027	3.5717	0.3575	0.9718
100	0.0019	2.5081	0.3003	0.9773

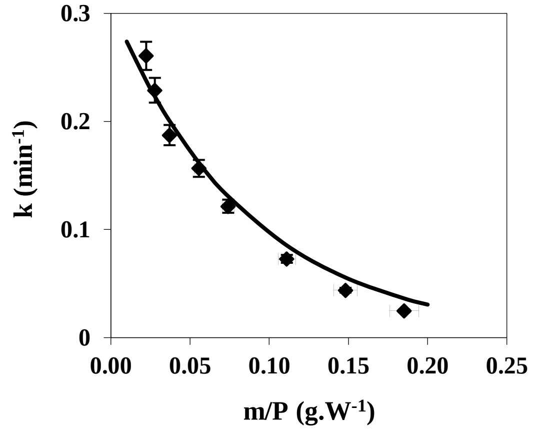

Figure 3: The relationship between the values of drying rate constant (*k*) versus sample amount/power (*m/P*), ■ Experimental data, — model.

output power to sample amount, Arrhenius type exponential model, Eq. (5), which was derived by Dadali *et al.* (2007c) was used with the standard error (σ) of $0.81x10^{-11}$ and coefficient of determination of (R^2) statistical value of 0.9761. The fitness of the data with the model was illustrated in Fig. 4.

$$D_{eff} = D_o \exp\left(\frac{-E_a .m}{P}\right) \quad (5)$$

The values of D_o and E_a were estimated as $1.421x10^{-10}$ m^2/s and 11.12 W/g. As a conclusion, the value of E_a found from this study was quite similar to the value (11.54 W/g) obtained from the previous section by using the Eq. (4).

F. The relationship between drying rate constant and effective moisture diffusivity

For prediction of the relationship between drying rate constant and effective moisture diffusivity, Eq. (6) was derived by using Eqs. (4 and 5) with the assumption of E_a values were quite similar to each other as mentioned in the previous section. The theoretical values of drying rate constant, k_{th}, obtained from Eq.(4) and the theoretical values of effective moisture diffusivity $(D_{eff})_{th}$ obtained from Eq. (5) were fitted sufficiently to Eq. (6) with the standard error (σ) of 0.0019 and the coefficient of determination of (R^2) statistical value of 0.9996. The value of constant (*A*) was obtained as $2.13x10^9$ $min^{-1}.m^{-2}.s$. The fitness of the data with Eq. (6) was illustrated in Fig. 5.

$$k_{th} = A.(D_{eff})_{th} \quad (6)$$

IV. CONCLUSIONS

Drying kinetics of tea leaves was investigated at various microwave output powers and sample amounts in a microwave oven. Drying time decreased considerably with increase in microwave output power and with decrease in sample amount of tea leaves as well by using microwave drying technique.

Among of eight models used in this study, the Page model provided a good agreement between experimental and predicted moisture ratio values with higher coefficient of determinations and lower standard error of estimates. The value of the drying rate constant, k, increased with the increase in microwave output power, on the other hand, decreased with the increase in sample amount. The effective moisture diffusivities increased with the increase in microwave output power. On the other hand, the values of the effective moisture diffusivities decreased, as the sample amount increased.

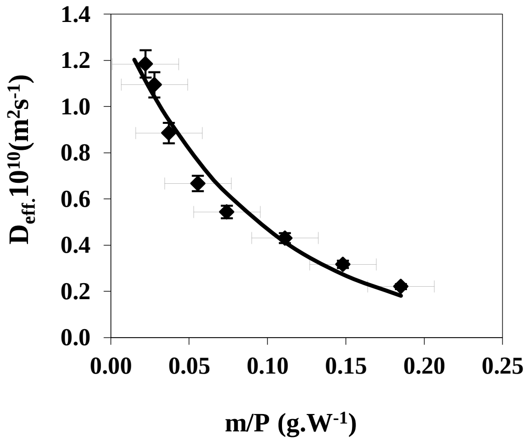

Figure 4. The relationship between the values of effective moisture diffusivity (D_{eff}) versus sample amount/power (*m/P*), ■ Experimental data, — model.

On the other hand, the activation energy of tea leaves was calculated by using the exponential expression based on Arrhenius equation and found similar as 11.54 and 11.12 $W.g^{-1}$, respectively. The benefit of this study, by using the exponential equation based on Arrhenius equation, k_{th} values for drying kinetics of tea leaves can be calculated by choosing the microwave output power and sample amount as well as for calculation of $(D_{eff})_{th}$ value.

It should be noted that the results obtained are limited with the microwave oven used in this study; because, any microwave oven exhibit individual power distribution. Though, the study performed demonstrates the methodology and guidance for use of microwave oven as gentle tool for drying of heat sensitive substances.

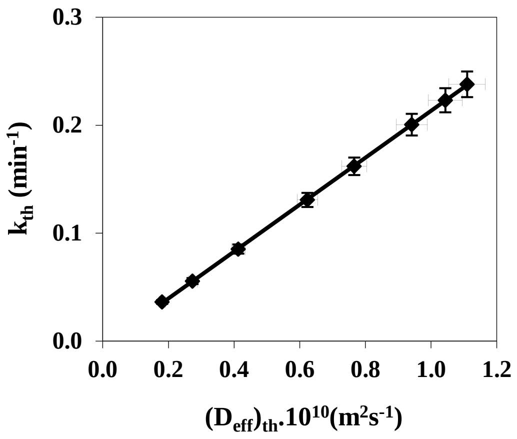

Figure 5. The relationship between the theoretical values of drying rate constant (k_{th}) and the theoretical values of effective moisture diffusivity $(D_{eff})_{th}$; ■ Theoretical data, — model.

NOMENCLATURE

E_a	Activation energy (W/g)
D_{eff}	Effective moisture diffusivity (m^2/s)
D_0	Pre-exponential factor (m^2/s)
k	Drying rate constant obtained by using Page model (min^{-1})
k_0	Pre-exponential constant (min^{-1}),
L	Half of thickness of the samples (m)
m	Mass of raw sample (g)
M_t	Moisture content at a specific time (g water/g dry base)
M_0	Initial moisture content (g water/g dry base)
M_e	Equilibrium moisture content (g water/g dry base)
MR	Moisture ratio
P	Microwave output power (W)
t	Drying time (min)

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