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Electrohydrodynamic, oven and natural drying of mint leaves and effects on the physiochemical indices of the leaves

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Abstract: Electrohydrodynamics (EHD) enhanced the thin-layer drying of mint (*Mentha Spicata* Huds) leaves using multiple point-to-plate electrodes. Its configurations were carried out under DC high voltage and its effects were compared to oven drying at 40 °C and open-air-drying with natural convection at 25 °C. The EHD was run in an optimum electric field of 3.2 kV·cm^{-1} using positive ionising conical needles to a plate electrode covered by a steel screen grid to prevent the leaves drifting. The samples subjected to the EHD and oven exhibited faster drying kinetics than natural convection. Compared to the oven drying, the EHD electrical power consumption was negligible. The EHD method developed fewer undesirable changes in the colour features and the leaves' total chlorophyll, whereas the oven-dried sample colour underwent a significant change in colour. The samples dried by EHD had lower active microorganisms. The empirical modelling, based on the maximum value of R^2 and the minimum value of RMSE and SSE between the experimental and predicted moisture ratios, showed that the diffusion and logarithmic models were the best models for describing the EHD and oven drying behaviour of the mint leaves.

Keywords: corona; EHD; modelling; thin layer

Most herbs and spices are marketed in dry form since they contain a high percentage of water and can suffer alterations in their composition. Mint (*Mentha spicata* Huds) is a genus of the Labiate family, which comprises a full number of species, varieties, and hybrids.

Food drying is widely applied for food preservation and may lead to a significant decrease in the weight and volume, minimising the packaging, warehousing, and transportation costs (Kavak-Akpinar 2010). Most of the conventional thermal treatments, such as hot air or sun drying leads to the undesirable thermal degradation of the finished wares. A high temperature is the one factor associated with the potential adverse nutritional changes in the dried product (Bajgai et al. 2006). It is essential to dry such products at desirably low temperatures to preserve their nutritional value.

A novel, innovative drying technique is based on the generation of a large number of ions by a corona discharge in a strong electric field. Air ions are produced by a corona discharge using a single or multiple point-to-plate electrodes. Due to the momentum transfer from the ions to the neutral air molecules, a corona wind flow is created. Corona wind turbulence and its vortex motion enhance the moisture transfer rate of water. EHD (electrohydrodynamics) improves the moisture transfer rates with a higher desirable product quality at lower energy costs (Alemrajabi et al. 2012). HPLC (high-performance liquid chromatography) analyses showed no foreign substances formed in the EHD-dried apple slices, which retained lighter colours with less browning than in the oven or with natural convection air (Hashinaga et al. 1999). Bajgai et al. (2006) found that EHD dries the produce and creates superior quality food products of high nutritional value as well as retaining the natural colour and textural characteristics. Lai and Sharma (2005) conducted a series of experiments to investigate the effect of the electrode geometry and showed that the performance of the multiple-needle electrode was better compared to wire and single-needle electrode configurations. In addition, a comparison of different polarities revealed that a positive corona discharge was more effective than the negative one at lower applied voltages. Alemrajabi et al. (2012) found that positive corona drying consumed less energy than negative corona drying. Ethylene oxide, ozone, irradiation (gamma, plasma, infrared, UV – ultraviolet), and superheated steam can all be used for the bacterial reduction in the spice industry. Irradiation methods are valid and the scientific theory is viewed as being safe (Leistritz 1997). EHD causes a decreasing number of bacteria with an increasing electric field power and an increasing corona exposure time. Electroporation occurs because the cell membrane has a specific dielectric strength, which can be exceeded by the EHD electric field (Al-Hilphy 2014).

Although many studies have been conducted on the drying characteristics of mint leaves using different techniques, no comprehensive research was found on EHD drying. The aim of the present research was to compare the rate of moisture loss, the energy consumption, and a colour quality assessment of the EHD, oven and natural drying methods for mint leaves and to find the best prediction model (Ozbek, Dadali 2007; Kavak-Akpinar 2010) for describing the corresponding drying behaviour.

MATERIAL AND METHODS

Fresh, leaf samples were obtained from plants at the Isfahan University of Technology Research Greenhouse. They were washed and stored at 4 ± 0.5 °C in a refrigerator for about one day for the moisture calibration. Before the drying experiments, the samples were taken out of the refrigerator, and the leaves were separated from the stems and then weighed. Four samples of the leaf, each weighing 3 g, were dried in an oven at 70 ± 1 °C for 24 h to determine the initial moisture content (AOAC 2019). The initial moisture content was calculated 0.75 g : g (wet weight) for the mint leaves. Samples were randomly chosen from the pile and spread uniformly in a single layer with three replications.

The EHD setup consists of a vertically mounted multiple sharp conical needle electrode 0.2 mm in diameter at the point projected to a fixed horizontal grounded metallic plate on which the samples to be dried were placed. The distance between the electrodes and the grounded plate electrode was adjustable (Figure 1). The sharp points were connected to a direct current power source that supplied a positive pole (EHD$^+$) of a high voltage DC power supply (PNC 40000-5, Heinzinger electric GmbH, Germany) (Zheng et al. 2011; Alemrajabi et al. 2012). The samples were spread uniformly in a single layer on the grounded plate electrode on an aluminium dish, covered by a fragile steel screen grid to prevent the leaves drifting into the needle cathode.

The preliminary study showed that a plastic screen grid had a slower drying rate, and the steel grid was better. A set of initial experiments with three-electrode gaps (30, 40, and 50 mm) and three electric voltages (13, 14.5, and 16 kV) were performed to determine the optimum electric field strength for the EHD drying rate. These domains were based on the limitation of the corona discharge setup configuration, while sparks occurred at the particular electrode gaps. The oven drying was carried out at 40 °C in an oven with forced circulation. Therefore, three different methods were used to dry leaves, namely, natural convection in the shade, oven drying, and EHD drying. The samples were weighted rapidly at regular time intervals using an electronic digital balance (accuracy 0.01 g, model GF-400, A&D, Japan) to determine the drying rate. All the experiments were carried out in an ambient temperature (25 ± 1 °C) with a relative humidity of 0.25 g : g, and it took 6 hours. The surface temperature of the mint leaves during the first hours of the EHD drying was measured during the experiments using thermometer

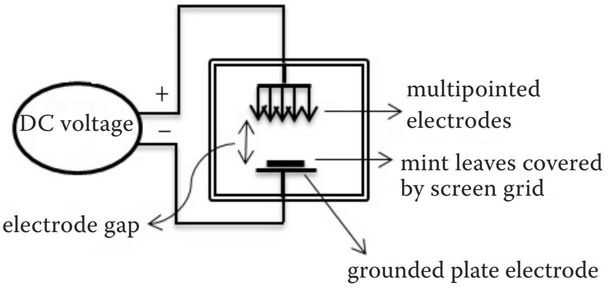

Figure 1. A schematic of the multiple pointed electrodes-to-plate used for the EHD drying of mint leaves covered by a steel screen grid to prevent the leaves drifting

(MLX 90601 IR, Melexis, Germany) with an accuracy of 0.1 °C. The leaves were cooled to room temperature for 10 min after the drying treatment and kept in air-glass jars. The microorganisms of the fresh and dried leaves were determined for the bacterial colony count (CFU·g^{-1}) according to ISO 4833-1:2013. The experiments were independently performed three times, and a t-test was used to compare them. The consumed energy (E) per unit reduced mass run on the full capacity of each drying technique was determined using Equation (1):

$$E_{\mathrm{c}} = \frac{V \times I \times t \cos \varnothing}{m_{\mathrm{i}} - m_{\mathrm{f}}} \quad (1)$$

where: E_{c} – the consumed energy (J·g^{-1}), V – the voltage of the power supply (V), I – the consumed electrical current (A), Φ – the angle of phase variance (deg.); m_{i} and m_{f} – the initial and final mass values of the samples (g) at the time interval t (s), respectively.

Colour and chlorophyll measurement. When a colour is expressed in CIELab, L^* defines the lightness, a^* denotes the red/green value, and b^* denotes the yellow/blue value (Rathore, Kumar 2012). The colour coordinates (L^*, a^*, and b^*) of the mint leaves were carried out three times before and after drying using a colour difference meter (Texflash DC 3881, Switzerland) spectrophotometer. Given Δa^*, $\Delta b^*$and ΔL^*, the total difference or distance on the CIELab diagram can be stated as a single value, known as ΔE using Equation (2) (the subscript '0' designates fresh samples).

$$\Delta E = \left(\Delta a^{*2} + \Delta b^{*2} + \Delta L^{*2}\right)^{1/2} \quad (2)$$

where: $\Delta a^* = a^* - a_0$, $\Delta b^* = b^* - b_0$, $\Delta L^* = L^* - L_0$

The chroma (C) showed colour saturation, which is proportional to the colour intensity. The hue angle (h) is another feature used to characterise the colour in food products. An angle of 0 or 360 degrees represents a red hue, and 90, 180, 270 degrees represent yellow, green, and blue ones, respectively. The hue angle and chroma were calculated as follows (Equations 3 and 4):

$$h = tan^{(-1)}\left(\frac{a^*}{b}\right) \quad (3)$$

$$C = \left(a^{*2} + b^{*2}\right)^{1/2} \quad (4)$$

where: a, b – constants.

Table 1. The empirical models used to predict the moisture ratio of the mint leaves during the thin layer drying by the EHD and oven drying methods

Model name	Equation
Page	$\mathrm{MR} = \exp(-kt^{\mathrm{n}})$
Logarithmic	$\mathrm{MR} = a \times \exp(-kt) + c$
Modified Henderson & Pabis	$\mathrm{MR} = a \times \exp(-k_1 t) + b \exp(-k_2 t) + c \exp(-k_3 t)$
Diffusion	$\mathrm{MR} = a \times \exp(-kt) + (1 - a) \exp(-kbt)$

MR – moisture ratio; k – drying constant; t – duration of the experiment; a, b and c – constants

A chlorophyll meter CL-01 (Hansatech, UK) was used to measure the total chlorophyll content of the mint leaves over the three replications. The CL-01 chlorophyll content meter provides a convenient, low-cost method of measuring the relative total chlorophyll content of the leaf samples. The device determines the relative total chlorophyll content using a dual-wavelength optical absorbance (620 and 940 nm wavelength) of the leaf samples. The relative total chlorophyll content is displayed in the range of 0–2 000 units (Cassol et al. 2008).

Empirical modelling of the drying kinetics. Many factors determine the application of the drying models for a specific product include the drying condition, shape, dimension, and initial moisture content. However, the model based on Newton's law is the most unaffected complexity compared to exponential models that are derived from Fick's second law of diffusion. Therefore, the experimental drying curves were fitted to the four different models, as shown in Table 1. The non-linear regression analyses were performed using Matlab software (version 7.6). The terms used to evaluate the goodness of the fit were the coefficient of determination (R^2), the RMSE (root mean square error) and SSE (sum of squares error) between the experimental and predicted moisture ratio values. The higher the R^2 amount and the lower the RMSE and SSE values are, the better the goodness of the fit is. The values of R^2, RMSE, and SSE were calculated using the following Equations (5–7):

$$R^2 = \frac{\sum_{i=1}^{n} (MR_{\mathrm{pre,i}} - \overline{MR})^2}{\sum_{i=1}^{n} (MR_{\mathrm{exp,i}} - \overline{MR})^2} \quad (5)$$

where: R^2 – the coefficient of determination; MR_{pre} – predicted moisture ratio; MR_{exp} – experimented moisture ratio.

$$\mathrm{RMSE} = \left[\frac{1}{n}\sum\nolimits_{\mathrm{i}=1}^{n}\left(MR_{\mathrm{exp,i}} - MR_{\mathrm{pre,i}}\right)^{2}\right]^{\frac{1}{2}} \tag{6}$$

for more explanation see Equation (5).

$$\mathrm{SSE} = \left[\sum\nolimits_{\mathrm{i}=1}^{n}\left(MR_{\mathrm{exp,i}} - MR_{\mathrm{pre,i}}\right)\right]^{2} \tag{5}$$

where: SSE – sum of squares error; for more explanation see Equation (5).

RESULTS AND DISCUSSION

Based on the preliminary experiments and the EHD setup limitation, such as the power supply capacity, resistance circuit, and its current amperes, a suitable voltage and electrode gap was selected to prevent its arc and spark on the mint leaves. However, three-electrode gaps (30, 40, and 50 mm) and three electric voltages (13, 14.5, and 16 kV) were used, and the optimum electric field strength of 3.2 kV·cm^{-1} (16 kV at 50 mm electrode gap) was determined. The electric field strength of 3.2 kV·cm^{-1} led to the maximum moisture loss, which was applied in the remaining EHD drying experiments (Figure 2).

The effect of the electric field strength on the leaves' moisture loss was investigated for 45 min drying periods during the preliminary tests. Other researchers have also reported a similar sigmoid trend at a different electric field strength (Kirschvink-Kobayashi, Kirschvink 1986). The samples were monitored for their temperature variations throughout all of the preliminary EHD drying tests. During the first hours of drying, the evaporation rate was high, and the latent heat of evaporation was partially supplied by the samples, leading to a decrease in the temperature. Reportedly, the surface temperature of carrot slices never exceeded the natural convection temperature in EHD drying (Alemrajabi et al. 2012). On the other hand, oven-dried samples are exposed to a high temperature, which may adversely affect the product quality (Hashinaga et al. 1999). Hence, the non-thermal drying of EHD can be considered as an advantage over conventional drying methods. The variation in the moisture content of the mint leaves with time is shown in Figure 3 for the EHD, oven, and natural convection air-drying methods.

Although the required drying time for EHD can be quite long, the drying took place in a closed area to prevent contamination with dust, soil, sand particles, and insects. According to Figure 4, a constant rate period was not observed in the EHD and oven

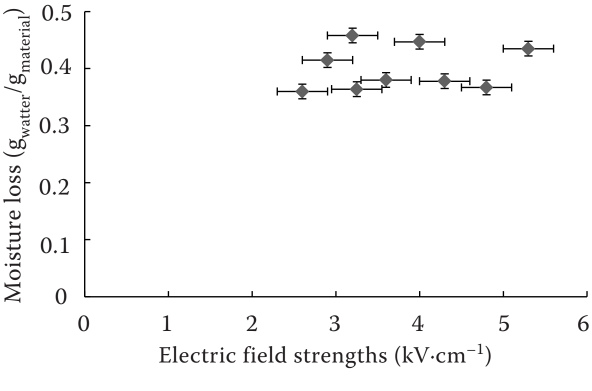

Figure 2. Variation of the moisture loss of the mint leaves at the different electric field strengths using a standard error.

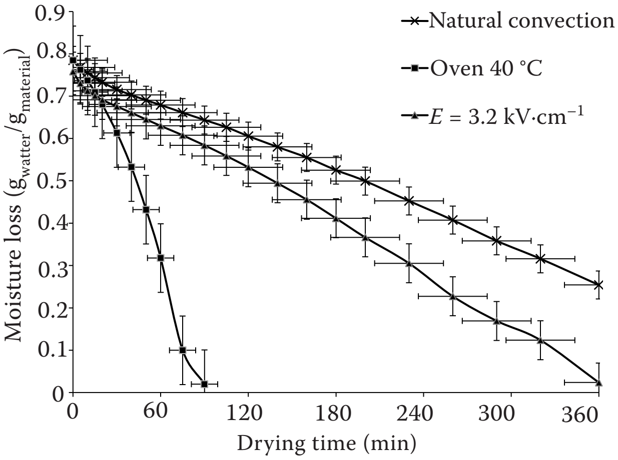

Figure 3. Variation in the moisture content with time of the mint leaves for the different drying methods using a standard error

E – electric field strength

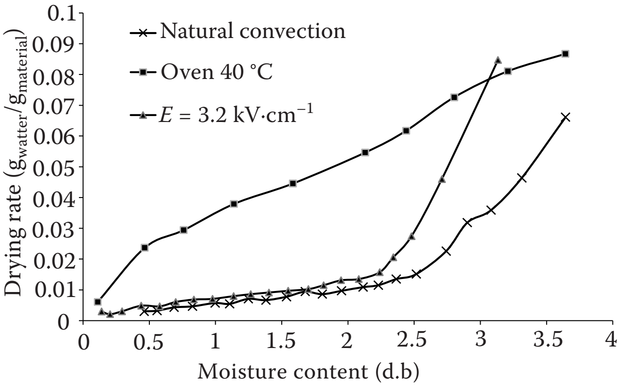

Figure 4. Variation in the drying rate with the moisture content of the mint leaves for the different drying methods

d.b. – dry basis

Table 2. The mean comparison of the colour features of the mint leaves dried by the different drying methods

Drying method	Temp. (°C)	L^*	a^*	b^*	ΔE	h	c
EHD (3.2 kV·cm^{-1})	25	$39.4^{b} \pm 0.7$	$-1.22^{b} \pm 0.2$	$14.7^{b} \pm 0.2$	12.1^{b}	$92.5^{b} \pm 0.1$	$14.7^{b} \pm 0.4$
Oven	40	$38.6^{b} \pm 0.3$	$-0.96^{c} \pm 0.2$	$12.2^{c} \pm 0.3$	14^{a}	$94.3^{b} \pm 0.3$	$12.6^{c} \pm 0.2$
Natural convection	25	37.5 ± 0.2	-1.69 ± 0.1	13.2 ± 0.1	13.8	95.5 ± 0.1	13.3 ± 0.2
Fresh mint	–	47.0 ± 0.3	-9.09 ± 0.2	19.9 ± 0.3	–	114.2 ± 0.2	21.9 ± 0.3

$^{a-c}$the means in the same column followed by different superscripts are significantly different (*t*-test); L^* – the lightness; a^*– the red/green value; b^*– the yellow/blue value; ΔE – net colour purity difference; h – hue angle; c – chroma

drying; however, the entire drying occurred in a falling rate period. During this period, the migration of moisture occurred through the mechanism of diffusion. The corona wind moved the humidity on the surface of the samples and reduced the external resistance to the moisture transfer. The falling rate period showed the diffusion was essential, and the internal resistance inside the sample nodes, because of the moisture gradients, was higher and an internal transfer was dominant.

Colour changes. The L^*, a^*, and b^* values in Table 2 revealed that the mint leaves' colour was affected in all the cases regardless of the drying method used. The L^* value decreased in all three drying methods, whereas the EHD least influenced it. This falling value indicates the samples were turning darker. High temperatures (hot air-drying) or a long drying time (EHD) could negatively affect the food colour because of enzymatic or non-enzymatic browning (Ohshima et al. 2007). Sharma and Prasad (2001) showed with an increase in the air-drying temperature, the sample colour became darker, implying more browning accrued. The discolouration of the samples during any drying procedure may be related to the pigment destruction, ascorbic acid browning, and non-enzymatic Maillard browning.

A negative a^* – value indicated greenness, while this parameter increased gradually during the drying. The oven-dried samples lost their greenness more. In addition, the b^* – values shifted towards lower values, indicating yellowness in the oven-dried samples. The chroma, which means the stability of yellow colour, decreased during drying, and closely followed the b^*– values. The hue angle values decreased in the samples subjected to each of the three drying methods. A lower ΔE denotes a smaller colour change in the EHD (Table 2). Compared with the oven-dried mint leaves, the EHD dried ones have been reported to show slight colour changes and to remain more similar in colour to fresh fruits (Hashinaga et al. 1999; Alemrajabi et al. 2011). In addition, the nature of EHD mechanism is not provided on the microbial activation which causes the colour changes. The impingement of the corona wind on wet materials can produce an impact high shear force, which may be thought to be the cause of the enzyme inactivation similar to ultra-sonication for the preservation of such produce. The enzymatic activity could be changed with an electric field treatment (Ohshima et al. 2007). Enzymatic degradation is achieved with long-term drying such as EHD; however, EHD had a lower ΔE. The degree of colour change was dependent on the drying temperature, drying time, and oxygen level. According to Table 2, the oven-dried samples converted more total chlorophyll. The high temperature could lead to the replacement of the total chlorophyll, thereby converting the chlorophylls to pheophytins; however, the impact on the colour change increased when the temperature increased (Maharaj, Sankat 1996).

Microorganism reduction. The drying experiments showed the total microorganisms of the EHD dried leaves were lower, but no differences were noted among the oven drying and natural convection methods (Table 3). Mizeraczyk et al. (2002) showed EHD is responsible for the ozone transport in the discharge region of the corona as in non-thermal plasma technology, therefore, ozone oxidation or electroporation of the cell membrane (Al-Hilphy 2014) may treat the mint leaves' microorganisms. Brodowska et al. (2015) stated that ozone has a potential effect on biologically active substances and could be used in food manufacturing as an alternative decontamination method.

Electrical power consumption. To select a suitable dryer, the energy consumption and quality of the dried samples are critical features. Oven drying is a more energy-intensive (Table 3) because the energy consumed in the oven drying is partly used to compensate for the heat loss from the oven to the

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Table 3. The consumed energy, colony count, and total chlorophyll difference of the mint leaves dried by the different drying methods (run on the full capacity of the dryers)

Drying method	Temperature (°C)	Electrical field (kV·cm^{-1})	Consumed energy (kJ·g^{-1})/capacity	Colony count (log CFU·g^{-1})	Chlorophyll converting
EHD	25	3.2	2.46^{b}	2.8^{b}	191^{b}
Oven	40	–	35.3^{a}	3.7^{a}	224.9^{a}
Natural convection	25	–	–	3.86^{a}	156.2
Fresh mint	25	–	–	4.08^{a}	7.99

CFU – coli forming unit

natural convection air. The electrical power in the EHD is far lower than oven drying. EHD consumed some electrical power to produce the corona wind (change-over of electrical to mechanical energy) and latent heat, the heat of vaporisation of water could be absorbed from the surrounding air (latent heat = 2.4425 kJ·g^{-1} water at 25 °C). From Table 3, the EHD consumed 2.46 kJ·g^{-1}, and the oven drying consumed 35.3 kJ·g^{-1} (run on the full capacity of the dryers) to dry the mint leaves. It may be concluded the energy consumed in the EHD drying is more efficient, whereas the different mechanisms in the EHD drying cause it to be efficient. Esehaghbeygi and Basiry (2011) have reported that EHD energy consumption for drying tomato slices at 3, 4 and 5 kV·cm^{-1} was 4.4–16.5 kJ·g^{-1}, while for oven drying of the same product at 55 °C was 200 kJ·g^{-1} run on the full capacity of each dryer. However, Kavak-Akpinar (2010) showed the energy utilisation ratio and improvement potential of mint leaves decreased with an increasing drying time and ambient temperature. In addition, microwave drying required a power intensity of 14.4 W·g^{-1} (Ozbek, Dadali 2007), whereas using microwave vacuum drying required 11.2 W·g^{-1} (Therdthai, Zhou 2009) for drying mint leaves. The microwave absorbed the energy to vaporise water and continuously increase the temperature; therefore, burn spots were found at the last stage of drying mint leaves (Therdthai, Zhou 2009).

Drying model. The evaluation of the drying kinetics as a function of the drying conditions could help us in the drying simulation to predict the suitable drying conditions. The model constants to fit the experimental moisture ratios to four thin layer models are shown in Table 4. Based on the R^2, RMSE, and SSE values, the diffusion and logarithmic models were considered the best for predicting the thin layer drying behavior of the mint leaves by EHD and oven drying, respectively.

As the drying was dominated by the falling drying rate period (according to Figure 4), the overall Fick's model performance over the whole drying period

Table 4. The model constants and statistical parameters used to model the EHD drying and the oven dryingof the mint leaves

Empirical model	R^2	RMSE	SSE	Model constants					
EHD drying									
Page	0.9844	0.3705	0.2745	$k = 0.2887$	$n = 0.7633$				
Logarithmic	0.987	0.3478	0.02299	$a = 0.9187$	$k = 0.00736$	$c = -0.036$			
Mod. Henderson & Pabis	0.9946	0.0243	0.00945	$a = 0.0692$	$b = 0.1165$	$c = 0.8496$	$k_1 = 0.212$	$k_2 = 0.21$	$k_3 = 0.0076$
Diffusion	0.9958	0.01968	0.0073	$a = 0.1538$	$b = 0.0295$	$k = 0.2551$			
Oven drying									
Page	0.996	0.0232	0.00486	$k = 0.0153$	$n = 1.192$				
Logarithmic	0.999	0.0105	0.00088	$a = 1.175$	$k = 0.022$	$c = -0.17$			
Mod. Henderson & Pabis	0.949	0.1104	0.06091	$a = 0.073$	$b = 0.247$	$c = 0.82$	$k_1 0.03$	$k_2 0.6$	$k_3 0.03$
Diffusion	0.993	0.0329	0.00868	$a = -19.19$	$b = 0.982$	$k = 0.04$			

R^2 – the coefficient of determination; RMSE – root mean square error; SSE – sum of squares error should be reasonably good. The variations in the experimental and predicted moisture ratios gained for the diffusion and logarithmic models for the EHD and oven drying, respectively, are presented in Figure 5. A good agreement was observed between the experimental moisture ratios and those predicted by the models. A more extended falling rate period showed that diffusion was governing the removal of the moisture from the samples. From Table 4, it can be shown that the models derived from Fick's second law could satisfactorily describe the drying curves of the mint leaves dried in the EHD. The *k* coefficients could be associated with the facility to remove moisture from the sample, and its values are related to the effective moisture diffusivity.

Sacilik et al. (2006) showed the diffusion model predicted a better fit to the experimental thin layer for the solar drying of organic tomatoes than the other models did. Similar results have been reported for the diffusion model of the apricot and fig (Togrul, Pehlivan 2004). In addition, the two-term and diffusion model represented the thin layer drying behaviour of mint leaves at 45 °C in a tunnel dryer (Kadam et al. 2011). On the contrary, Chen and Barthakur (1994) reported the EHD drying kinetics of potato slabs did not follow the Fickian diffusion model. Hashinaga et al. (1999) and Bai et al. (2011) recommended the polynomial equation for EHD drying of apple slices and fish. Ding et al. (2014) showed the Page model was the best mathematical model for EHD drying of cooked beef. These discrepancies can be due to the different experimental conditions.

The logarithmic model was considered to be the best for predicting the thin layer drying behaviour of mint leaves for the oven (Table 4). In addition, Doymaz (2006) showed logarithmic models had the best explanation for the thin-layer drying behaviour of mint leaves at a temperature range of 35–60 °C in a cabinet dryer. Similar results have been reported for the logarithmic model at a high-temperature range of 40–70 °C for corn ears and apple slices, respectively (Correa et al. 2011; Rayaguru, Routray 2012). However, the Page and modified Page models, as well as the Wang & Singh model, were the best to describe high-temperature drying in other research (Therdthai, Zhou 2009; Kavak-Akpinar 2010; Akonor, Amankwah 2012). Some widely used empirical models include a parabolic model (Wang & Singh), and Midilli et al. (2002) had undesirable convergence for drying mint leaves and their work did not fit our experimental data.

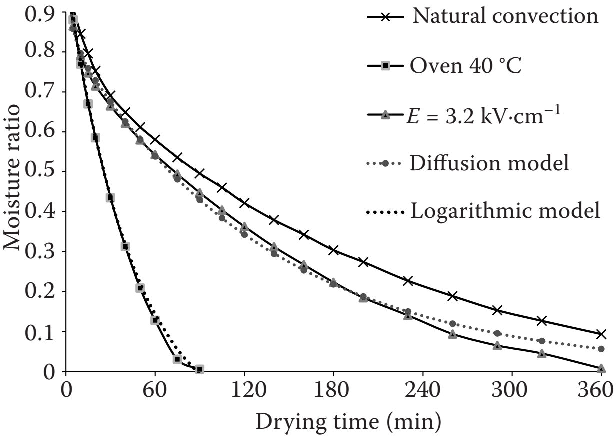

Figure 5. Variation in the moisture ratios with the time of the mint leaves for the different drying methods compared with the empirical diffusion and logarithmic model

CONCLUSION

The drying rate of mint leaves using the oven at 40 °C was rapid. No constant rate period was observed in the EHD drying. The lower air temperature in the EHD and its long drying time are the main features involved in the colour change during convective drying. The EHD affected the leaves' microorganisms. The energy consumption in the EHD drying was low, and its original colour remained almost intact, while considerable browning was observed in the leaves dried in the oven. The diffusion and logarithmic models were identified as the best prediction model for describing the EHD and oven drying behaviour of mint leaves, respectively. This work guides the optimisation and improvement in the mint leaves' drying efficiency by the EHD and oven drying method.

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