Industrial Crops & Products 123 (2018) 46–54

Contents lists available at ScienceDirect

Industrial Crops & Products

journal homepage: www.elsevier.com/locate/indcrop

Development and optimization of the new ultrasonic-infrared-vacuum dryer in drying *Kelussia odoratissima* and its comparison with conventional methods

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ARTICLE INFO

Keywords:
Ultrasonic
Infrared
Kelussia odoratissima
Medicinal plants

ABSTRACT

Among the post-harvest processing of medicinal plants, drying is an important and influential process. Given the numerous applications of medicinal plants, especially *Kelussia odoratissima*, in the food and pharmaceutical industries, the aim of this study was to compare the effects of the ultrasound-infrared radiation-vacuum method with conventional drying methods on the drying time, the total phenolic content (TPC), total flavonoid content (TFC) and antioxidant activity of *K. odoratissima*. ANOVA result showed that the effects of drying methods, drying temperature and their interaction effect on phenolic, flavonoid and antioxidant content were significant at 1% probability level. In the ultrasound-IR-vacuum method, by increasing temperature from 40 °C to 80 °C, the TFC increased by 35%. The highest antioxidant capacity was obtained for dry shade treatment, followed by dry sun treatment and three temperatures, i.e., 40 °C, 60 °C and 80 °C, in the combined method. The proposed optimal temperatures for the hot air, IR, and ultrasonic drying, were 63 °C, 66 °C and 71 °C, respectively.

1. Introduction

Aromatic and medicinal plants, as an important natural source, can be used as additives (Behruzian et al., 2017; Jamshidi-Kia et al., 2018) or natural antioxidants in the food and pharmaceutical industries due to their pharmacological properties, more safely than synthetic materials (Calín-Sánchez et al., 2011). In addition to the factors related to pre-harvest processing, which affect the quality of the active ingredients derived from medicinal plants, also post-harvest handling also affects medicinal plants' quality. (Hosseinzadeh et al., 2011; Omidbaigi, 2005). Besides the plants' inherent biochemical properties, harvest conditions, cultivation, geographical and environmental factors, and postharvest processes play a vital role in the quantity and quality of secondary metabolites, and consequently the biological and medicinal uses of medicinal plants and aromatic herbs (Hassanpouraghdam et al., 2010). The results of the studies on the qualitative characteristics of medicinal and culinary plants have demonstrated the effects of pre- and post-harvest factors, and in some cases, their interaction effects in this regard (Ippolito and Nigro, 2000; Subasinghe, 2011).

Among the post-harvest handling methods of medicinal plants, drying is an important and influential process. The main objective of the drying process is to reduce the water level to less than 15% to inhibit microbial growth and minimize biochemical changes, to preserve the characteristics of the color and aroma, and to maintain the active ingredients and the final products' quality (Tanko et al., 2005). Several studies have been conducted to investigate the effect of drying on medicinal plants in recent years. Modern technologies have also being seeking to minimize changes in plant quality (Orphanides et al., 2016). In addition, drying leads to a reduction in the weight and volume of the product, which facilitates its storage and transportation, and increases its shelf life. Several experimental studies on the effects of drying methods on the amount and constituents of the active ingredients of medicinal and aromatic plants have indicated that proper drying methods must be selected depending on the type of plant species, the type of active ingredient, and the type of plant tissue (storage location of the active ingredient), and to this end, the use of new methods and their comparison with the common drying methods of drying can be greatly helpful (Ahmadi et al., 2008; Azizi et al., 2009; Omidbaigi et al., 2004; Sefidkon et al., 2006).

Traditional and old methods such as natural drying (drying in shade and sun) and hot air drying are still the most important methods used for the production of dry plant material due to the use of minimal

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https://doi.org/10.1016/j.indcrop.2018.06.053
Received 10 March 2018; Received in revised form 1 June 2018; Accepted 12 June 2018

equipment and devices (Ebadi et al., 2011; Martinov and Oztekin, 2007). In the last few decades, drying in ovens at different temperatures has been observed to be effectively for a variety of plant materials (Hassanpouraghdam et al., 2010). Hot air dryers are very common, but the long drying time due to the low heat transfer rate to the internal parts of the foodstuff, which results in lower energy efficiency, and consequently elongated moisture reduction, a decrease in nutritional value due to the slowness of the process, and also shrinkage of the materials are some of the disadvantages of these devices (Azizi et al., 2009). In recent years, new drying methods have replaced traditional and old methods, some of which include freeze drying, microwave drying, infrared (IR) drying, and vacuum drying. IR dryers have flourished in recent years, and due to their low cost and simple and inexpensive equipment, have drawn comparatively more attention in developing countries (Mujumdar, 2014). Ultrasonication is one of the emerging technologies, that is being increasingly used in various industries and for processing purposes day by day. Ultrasonication is based on mechanical waves at frequencies above the human hearing range. High-energy ultrasonication is used as a pre-treatment before drying with hot air to improve the characteristics of the dried product by improving the transfer of mass and heat (Awad et al., 2012; Hosseinzadeh Samani et al., 2018; Samani et al., 2017).

The medicinal plants of Iran are diverse due to its favourable climate and geography. The Umbelliferae family includes approximately 275 genera and 2850 species. 8 *Kelussia* is one of the most recently identified genera of this family with only one species, *Kelussia odoratissima* Mozaff., which occurs only in Iran. This self-growing, sweet-smelling, monotypic medicinal plant occurs exclusively in a small area in west of Iran, where it is locally called *Karafse koohi*. The aerial part of the plant is routinely used as garnish and a natural sedative agent. Few studies have so far been conducted to address the pharmacological properties, such as antioxidant, anti-inflammatory, fibrinolytic, hypoglycemic, and analgesic properties of *K. odoratissima* (Ahmadi et al., 2007).

Given the numerous applications of medicinal plants, especially *K. odoratissima*, in the food and pharmaceutical industries, and because the vacuum-IR-ultrasonic combination drying method has not yet been investigated, the aim of this study was to investigate the effects of the proposed method in comparison with conventional methods of drying on the time and kinetics of drying of *K. odoratissima*, as well as on the total phenolic and flavonoid content and antioxidant activity of this plant.

2. Materials and methods

2.1. Plant material

Leaves of *K. odoratissima* at an early stage of growing were collected from the central Zagros region in west of Iran on Zard-Kuh Mountain in March 2015 (32.3297°N, 50.1112°E; 2100 m above sea level). Sample identification was done by expert botanists (Shirmardi, Hamzeh Ali, PhD., Research Center of Agriculture and Natural Resources, P.O. Box 415, Shahrekord, Iran). The voucher specimen (No. 207) was deposited at the Herbarium of Shahrekord University of Medical Sciences (SKUMS). The leaves were then transferred to the laboratory under standard conditions for drying, extraction and measurement of total phenolic content, total flavonoid content, and antioxidant activity.

2.2. Determining the initial moisture of samples

The initial moisture content of the *K. odoratissima* was determined according to the AOAC standard (Horwitz, 2002). Three samples of specific weight were selected from the plant and placed in an oven at 105 °C for 24 h and the moisture of the samples was calculated by Eqs. (1) and (2) (Hosseinzadeh et al., 2011; Martinov and Oztekin, 2007).

$$M_{d.b} = (W_w - W_d) / W_d \quad (1)$$

$$M_{w.b} = (W_w - W_d) / W_w \quad (2)$$

where:W_w: The initial weight of the plant material before drying;W_d: The weight of the plant material after 24 h of drying at 105 °C;$M_{w.b}$: The initial moisture content of the plant material based on fresh sample weight; and$M_{d.b}$: The Initial content of plant material based on dried sample weight.

It should be noted the initial and final moisture content (w.b) of all treatments were 73 ± 3.1% and 11 ± 1.4% respectively.

2.3. Drying methods

2.3.1. Structure of ultrasonic-IR-vacuum dryer

Sonication was carried out by an ultrasonic cleaner (7500s, Panasonic Co., Iran) with the power of 100 W, thermal power of 150 W, the frequency of 28k ± 5%Hz, the capacity of 6 L, and dimensions of 15 × 15 × 30 cm. The vacuum was created by a vacuum pump (Germany) with 15 mbar ultimate pressure and 22 L/min pump speed. The temperature was applied by IR lamp and the temperature (40 °C, 60 °C, and 80 °C) were adjusted by changing the height of IR lamp and measured with a thermocouple (Fig. 1).

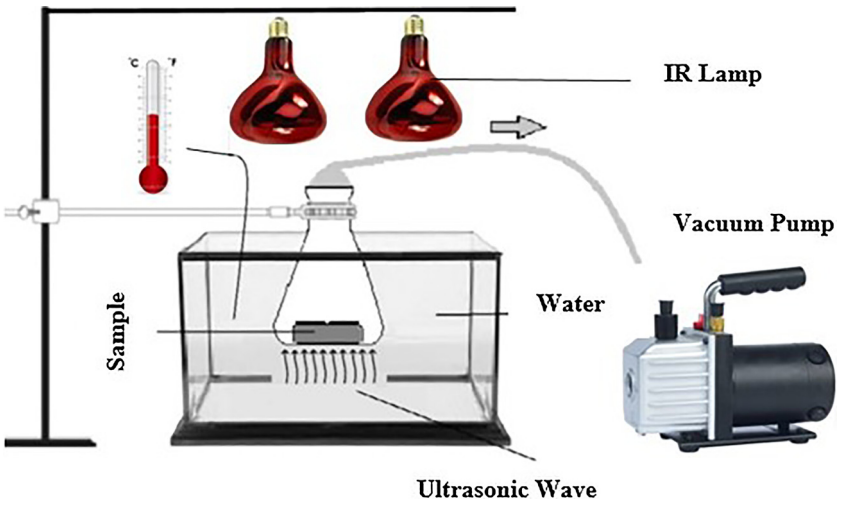

Fig. 1. Experimental setup of the ultrasonic-infrared-vacuum dryer.

2.3.2. Oven-drying method

In the oven-drying method, an oven was used for drying. Before starting the experiment, in order to achieve a stable state, the oven was lit for 1 h at the desired temperature and then the samples were dried. In this method, the samples were dried at three temperatures, 40 °C, 60 °C, and 80 °C, and the weights of the products were recorded in a time interval of 20 min as the drying was being performed (Pirbalouti et al., 2013).

2.3.3. IR-hot air dryer

To carry out the experiments, the IR dryer was developed and optimized in the University of Shahrekord, Shahrekord, Iran. This dryer performs drying by both hot air and IR. The dryer used in the current work can be set at an ambient temperature to 80 ± 2 °C. The air velocity in the dryer channel can be adjusted up to 1.5 m/s. The major components of the dryer include the dryer channel, in the middle of which the sample is placed on a tray, an exhaust blade for air flow within the channel, air heaters, channel valves to adjust the air velocity, IR heating lamps, IR chamber, balance, temperature control unit, and record unit of data. A chamber was used to set up an IR source. For the thermal insulation of the chamber, 2.5 cm thick wooden walls were used. The walls were covered with aluminum foil to minimize the absorption and to maximize the reflection of IR radiation. The chamber was built in a way that could adjust the distance between the IR source and the sample (e.g. 10, 15, 20, 25, 30, and 35 cm). Lamps (250 W, 220 V) used in the dryer were coated electric strings (Osram, Slovakia).

2.3.4. Comparison of drying methods

In this study, sun and shade drying methods (19–26 °C), as common methods used for drying most medicinal plants, as well as controlled oven-drying and IR drying methods were compared with the proposed ultrasonic-IR-vacuum drying method.

To dry *K. odoratissima* by shade drying method, an adequate amount of leaves was isolated from the fresh sample of the plant and dried at room temperature in the shade. To perform drying by dry sun drying, the plant was dried in the sun under the conditions in which the wind was gentle.

2.4. Preparation of the K. odoratissima extract

Different treated-dried *K. odoratissima* leaves were pulverized by powder by an electric grinder and then sieved (particle size: < 125 μm). The pulverized plant was macerated in ethanol: water (7:3) for 72 h are room temperature (3 times). Extraction fluids were filtrated and then concentrated using a rotary evaporator (EYELA, Japan) under decreased pressure at 38 °C. Finally, viscous extract was dried in an incubator (Memmert, Germany) at 37 °C to remove solvent completely. The obtained extracts were stored in a sealed vial at 4 °C until they were needed.

2.5. Phytochemical screening of ethanolic extracts

2.5.1. Measurement of total phenolic content (TPC)

The total phenolic content of treatments was measured by a colorimetric technique using the Folin-Ciocalteu reagent followed by a slightly modified method of Ainsworth (Ainsworth and Gillespie, 2007). Briefly, 0.5 mL of plant extract (100 μg/mL) was poured into a test tube and sufficiently mixed with 2 mL of a 1:10, deionized water-diluted Folin-Ciocalteu reagent and 2 mL of 7.5% Na2CO3 w/v. The reaction mixture was incubated at room temperature for 30 min as it was intermittently shaken so that color could emerge. Absorbance at 765 nm was measured using a UV–vis Spectrophotometer (Unico 2100). A standard curve of the assay was plotted by using gallic acid in 60% aqueous methanol. A mixture of water and reagents was used as blank. The data on total phenolic content were expressed as mg/g gallic acid equivalent (GAE) of dry extract.

2.5.2. Measurement of total flavonoid content (TFC)

The TFC of crude extract was measured using a modified aluminium chloride colorimetric method (Rahimi-Madiseh et al., 2017). Briefly, 50 μL of crude extract (1 mg/mL ethanol) was made up to 1 mL of methanol, mixed with 4 mL of distilled water and then 0.3 mL of 5% $NaNO_2$ solution; after a 5-min incubation, 0.3 mL of 10% $AlCl_3$ solution was added to the mixture, and then the resulting solution was left for 6 min. Then, 2 mL of 1 mol/L NaOH solution was added, and the volume of the mixture was increased to a final volume of 10 mL by addition of double-distilled water. The mixture was allowed to stand for 15 min, and the absorbance at 510 nm wavelength was recorded. The TFC was determined according to a calibration curve, and the results were presented in terms of mg rutin equivalent/g dry weight.

2.5.3. Determination of antioxidant activity by DPPH scavenging assay

The free radical scavenging activity of the treatments was studied by 1,1-diphenyl-2-picrylhydrazyl (DPPH) radical scavenging method, as described previously, with certain modifications (Moradi et al., 2017). The mixture of the assay contained 200 μL of 1.0 mmol/L DPPH radical solution in methanol and 100 μL of extract solution at different concentrations. The solution was quickly mixed and then incubated at 37 °C for 20 min in the dark. The reduction in the absorbance of each solution was calculated at 517 nm using UV/Vis spectrophotometer. DPPH radical solution with 100 μL methanol was used as blank. The percentage of radical scavenging (%) was calculated according to the formula below:

$$\%\textit{Free radicals scavenging activity} = \frac{A_c - A_s}{A_c} \times 100 \quad (3)$$

Where A_c = Absorbance of control at 517 nm; A_s = Absorbance of the sample.

The concentration of the sample that was needed to scavenge 50% of the DPPH free radical (IC_{50}) was calculated according to the curve of %inhibitions plotted against the corresponding concentration.

2.6. The analytical, optimization, and modelling method

2.6.1. Data analysis

Independent variables in this study were drying methods and temperature and the dependent variables were drying time, and phenolic, flavonoid, and antioxidant content. It should be noted that in the other part of the study, the optimization of different methods (final moisture contents was 11% w.b) would be investigated. The proposed experimental design is a randomized complete block design. Data analysis was performed using the Minitab software and mean values were compared by the LSD. $P < 0.05$ was considered significance level (Hemmatian et al., 2012).

2.6.2. Drying kinetics

Thin layer drying curve models for *Kelussia odoratissima* was selected based on 6 models recommended in previous studies, as shown in Table 1 (Evin, 2012). In most studies, drying kinetics model based on moisture ratio (MR) has been obtained. In these models, the moisture of

Table 1
Mathematical models given by various authors.

Equation	Model
MR = exp(−kt)	Lewis
MR = exp(−ktn)	Page
MR = exp[(−kt)n]	Modified Page
MR = a.exp(−kt)	Henderson & Pabis
MR = a.exp(−kt) + c	Logaritmic
MR = a/(1 + b.exp(kt))	Logistic

MR: moisture ratio, t: drying time, a,c,n, k_0, k_1: empirical constants in drying models.

the sample during drying is indirectly obtained (Evin, 2012). In this study, the MR changes to the drying time (t) of the sample were also plotted, and then, mathematical models were extracted by the MATLAB 2013 software. The MR was calculated by Eq. (4):

$$MR = \frac{M_t - M_e}{M_o - M_e} \quad (4)$$

Where, MR is moisture ratio (dimensionless), Mt is the moisture content at any moment, Mo is primary moisture content and Me is moisture balance, which is in terms of percentage and dry basis (d.b.). The amount of Me is small compared with Mo and Mt, therefore, by excluding Me, Eq. (5) is simplified as follows:

$$MR = \frac{M_t}{M_o} \quad (5)$$

Three criteria of coefficient of determination (R^2), chi-square (χ^2) and root mean square error (RMSE) was used to assess the best model. Thus, the model with higher values of R^2 and lower values of χ^2 and RMSE was considered as the best one.

$$\chi^2 = \frac{\sum_{i=1}^{n} (MR_{\exp,i} - MR_{pre,i})^2}{N - n} \quad (6)$$

$$RMSE = \left[\frac{1}{N} \sum_{i=1}^{n} (MR_{pre,i} - MR_{\exp,i}) \right]^{1/2} \quad (7)$$

Where, $MR_{exp,\ i}$ is the moisture ratio obtained during the ith measurements, MRpre, i is the predicted moisture ratio of the model in the ith measurements, N is the number of observations (data) and n is the number of the constants used in the equation. The coefficients and constants of the models in Table 1 were calculated at different temperatures using regression.

2.6.3. The method of optimizing and modelling of response surface method

Response surface method comprises a set of mathematical and statistical techniques that are used to analyze, model, and optimize processes where the rate of interest has already been affected by many variables. Therefore, implementing this method in the laboratory for all treatments is faced with time constraints and requires stupendous costs (Samani et al., 2016).

The use of the response surface method to determine the relationship between the response and the independent variables in a research, includes the steps as follows: Selecting the independent test variables and determining the range of each variable; drawing and verifying the equation model; and drawing the response curves, performing analysis of variance (ANOVA), and finally, determining the optimal point (Hosseinzadeh Samani et al., 2015). In order to obtain the model equation and determine the target function for optimization, the regression Eq. (8) was solved.

$$Y_i = \beta_\circ + \sum \beta i X i + \sum \beta_{ij} X_i X_j + \sum \beta_{ii} X_i^2 + \varepsilon \quad (8)$$

where

βo, βj, β_{ij}, and β_{jj} are constant coefficients, x_i and x*j* are independent variables in the process, and ε is random error.

3. Results and discussion

The moisture removal of the leaves was calculated in terms of the percentage difference between the different drying processes. The leaves of *K. odoratissima* was dried by different methods and then phytochemical content of the resulting extract was measured. The analysis of variance showed that drying, drying temperature, and interaction of these two factors on phenolic, flavonoid and antioxidant content were significant at 1% probability level (Tables 2–4). With regards to the sum of squared errors, the most effective factors on the phenolic, flavonoid, and antioxidant content, according to the ANOVA were the drying method, temperature, and drying method, respectively.

Table 2
Analysis of variance of drying methods and temperature on total Phenol content.

Source	DF	Sum of Square	Mean of Square	F
Drying methods	2	40.8822	20.4411	579.65**
Temperature	2	10.2351	5.1175	145.12**
Drying methods × Temperature	4	11.6162	2.904	82.35**
Error	18	0.6348	0.0353	
Total	26	63.3682		

** showed a significant effect at 1% level.

Table 3
Analysis of variance of drying methods and temperature on total flavonoid content.

Source	DF	Sum of Square	Mean of Square	F
Drying methods	2	6.199	3.099	1509.36**
Temperature	2	39.862	19.931	9706.23**
Drying methods × Temperature	4	61.279	15.32	7460.54**
Error	18	0.037	0.002	
Total	26	107.378		

** showed significant effect at 1% level.

Table 4
Analysis of variance of drying methods and temperature on antioxidant activity.

Source	DF	Sum of Square	Mean of Square	F
Drying methods	2	2881.48	1440.74	1382.07**
Temperature	2	568.92	284.46	272.88**
Drying methods × Temperature	4	234.08	58.52	56.14**
Error	18	18.76	1.04	
Total	26	3703.25		

** showed a significant effect at 1% level.

3.1. Effect of different drying methods and drying temperature on the total phenolic content and the flavonoid content of K. odoratissima

The results on the interaction effect between temperature and method of drying on the total flavonoid content showed that with increasing temperature in both hot air and IR drying methods from 40 °C to 60 °C, total flavonoid content increased, when the temperature reach to 80 °C, the TFC was reduced by 21% and 24% in hot air and IR drying methods, respectively (Fig. 2).

In the ultrasound-IR-vacuum combination method, when the temperature increased from 40 °C to 80 °C, the TFC increased by 35%. Comparison of the mean values of the treatments in the combination method showed that there was no significant difference between 60 °C and 80 °C at 5% level. In this regard, Dong et al. (2011) reported that as for TPC, temperature lower than that displayed high amount of TFC did not allow enough disruption of plant cell wall polymers causing better extracted; however, higher than optimal temperature cause to the decrease of TFC (Dong et al., 2011).

As Mrad et al. (2012) observed, a decrease in TFC during drying can occur as a result of polyphenols' binding to other compounds (proteins), or because of changes in the chemical structure of polyphenols that cannot be isolated or identified by available methods (Mrad et al., 2012). de Ancos et al. (2000) have proposed that polyphenolic compounds are likely to be deteriorated for numerous reasons other than

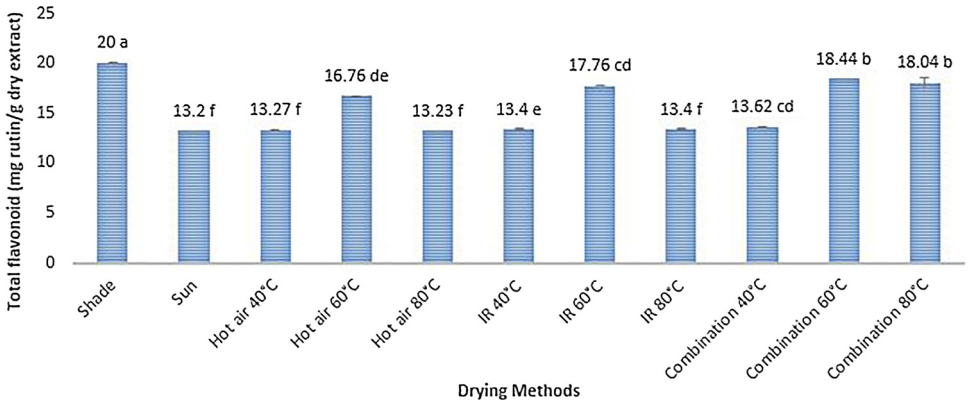

Fig. 2. Comparison of Total flavonoid (mg rutin/g dry extract) of different drying methods. Different letters showed a significant effect at 5% level.

heat treatment. These reasons include organic acid content, polyphenol oxidase activity, sugar concentration, and pH (de Ancos et al., 2000).

The effect of increasing the temperature on the TPC is similar to that on the TFC. With increasing temperature from 40 °C to 60 °C in both hot air and IR drying, the TPC increased by 2.5% and 5%, respectively, and when the temperature was increased to 80 °C, the TPC decreased by 4% and 3.5%, respectively. In the combined method, there was a direct correlation between the TPC and the temperature range. With increasing temperature from 40 °C to 60 °C, the TPC increased by 6%. Since the temperature of 80 °C was lower than the drying temperature of the *K. odoratissima* sample, the changes in the TFC and TPC were not significant at 60 °C temperature (Fig. 3). According to Fig. 2, when the temperature increases, the drying time in all three methods, i.e., hot air, IR, and combination, decreases. The maximum drying time, as with other studies (Hihat et al., 2017), was obtained for the shade drying method (3 days), but among the treatments investigated in this study, the highest drying time was obtained for the hot air drying at 40 °C (12 h) and the lowest drying time for the ultrasound-IR-vacuum method at 80 °C (4.6 h) (Fig. 4).

As a result, the total drying time significantly decreased as temperature increased. Consistently, Ahmed et al. (2001), in a study of coriander leaves drying with a cabinet dryer at 45, 50, 55, 60, and 65 °C, obtained similar results (Ahmed et al., 2001); however, Hihat et al. (2017) reported that the time of drying was 10 h for coriander leaves that have been dried in fixed bed dryer. It can be argued that increasing temperature decreases drying time considerably (Hihat et al., 2017).

The IR drying method reduces the drying time and increases the energy consumption and the quality of dried products due to consistent heating (Sellami et al., 2011). Samani et al. (2017) showed that for drying *Satureja bachtiarica*, in both the hot air and IR drying methods, the speed of the drying process increased, but in the IR drying method, compared to the hot air drying, the drying time decreased, drying efficiency increased, and the products' quality improved (Samani et al., 2017).

It seems that due to the reduction of drying time with increasing the temperature from 40 °C to 80 °C in the IR method, in contrast to the other two methods, the TPC increased by 6%. Indeed, the damage, caused by the increase in temperature, to the structure of phenolic compounds and flavonoids has not been effective over comparatively shorter period of time. Some studies have shown that the loss of macromolecules such as flavonoids in heat treatment can be attributed to the difficulties of drying conditions, especially the duration and temperature (Aghakhani et al., 2018; Schieber et al., 2001). Some researchers have reported the positive effects of heat and oven treatment on the TPC of plant material at temperatures above 80 °C, so that phenolic compounds formed at a high temperature (90 °C) due to the provision of phenolic compounds precursors along with non-enzymatic exchanges between the molecules (Hossain et al., 2010).

Among six plants from the Lamiaceae family, the highest TPC has been to exist in the natural method among other methods, such as drying in a natural manner at room temperature in the vacuum and the freeze-drying method (Hossain et al., 2010). In shade-dried *Dracocephalum moldavica* L., when compared to the sun-dried plant and the plant dried at different temperatures, the oven caused no change in phenolic compounds (Mohtashami et al., 2012). The increase in the TFC at 80 °C occurs because of the inactivation of the enzymes that are effective in decomposing and degrading flavonoids, as the oven at 40 °C decreases the TFC because of the optimal temperature of these enzymes. Highly similar observations have also been reported for

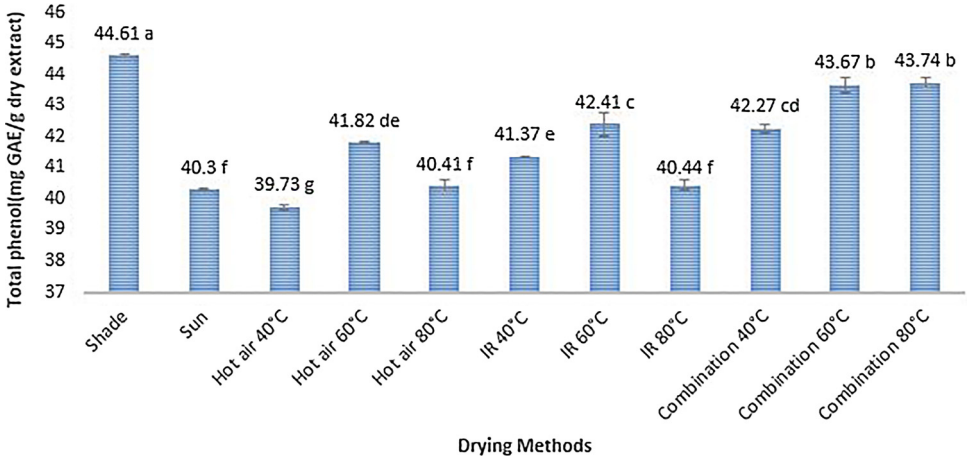

Fig. 3. Comparison of total phenol (mg GAE/g dry extract) of different drying methods. Different letters showed a significant effect at 5% level.

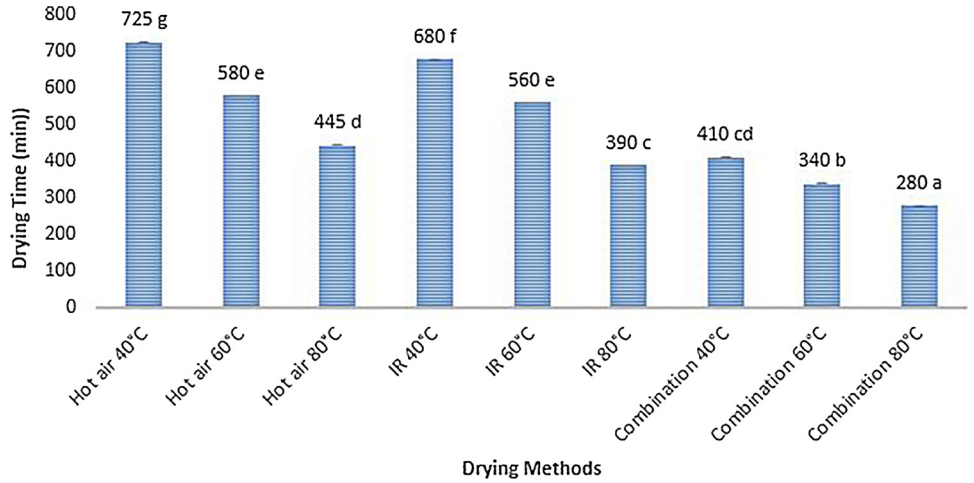

Fig. 4. Comparison of drying time (min) of different drying methods. Different letters showed a significant effect at 5% level.

quercetin (Rohn et al., 2007).

3.2. The effect of drying methods on DPPH radical scavenging capacity

The antioxidant capacity of treatments was measured by the DPPH assay. The DPPH assay has been extensively used to study the free radical scavenging activity of different antioxidant compounds. In the DPPH assay, the antioxidants that are able to donate hydrogen can reduce stable DPPH radical to the yellow colored, non-radical form of DPPH-H (Karimi et al., 2016). As Fig. 5 illustrates, the highest antioxidant capacity was obtained for dry shade treatment, followed by dry sun treatment and three temperatures, i.e., 40, 60 and 80 °C, in the combined method.

In all three methods, with increasing temperature, antioxidant capacity increased. In both hot air and IR drying methods, there was no significant difference between 60 °C and 80 °C temperatures at 5% *p* level, but in the combined method, the antioxidant capacity increased significantly at a *p* level of 5% among the three temperatures, 40, 60 and 80 °C. Our results are in agreement with those reported by Hamrouni-Sellami et al. (2013) who observed that the highest radical scavenging activity of the plant was obtained at 65 °C (Hamrouni-Sellami et al., 2013). In addition, 120 °C drying temperature might be due to generation and accumulation of Maillard derived melanoidins, which exert various levels of antioxidant activity and enhance antioxidant effects (Miranda et al., 2009). Hihat et al. (2017) also reported that at high temperatures (80 °C or 90 °C), dehydration caused a higher antioxidant activity compared to low temperatures such as 50, 60, or 70 °C. Taken together, the generation and accumulation of compounds with various levels of antioxidant activity during food dehydration can lead to antagonistic or synergistic effects between themselves or with other ingredients. These complex chemical interactions that affect food functional properties during drying are still being investigated (Hihat et al., 2017).

According to the dendrogram (Fig. 6), the least similarity was observed to be between the shade drying method and other methods. By dividing the 11 treatments into three clusters, it was observed that the dry sun, combined (80 °C and 60 °C), and IR (80 °C and 60 °C) drying methods were assigned to a cluster and the dry hot (40, 60, and 80 °C), IR, and combined (40 °C) methods to the other cluster.

3.3. The results of fitting of the models in different testing conditions

Thin layer moisture for *Kelussia odoratissima* during the drying process in each method was measured every 10 min. The models of Table 1 were fitted with the data obtained from the amounts of moisture ratio in the hot air, infrared and combination drying methods (under three temperatures of 40, 60 and 80 °C). Then, different thin layer drying models with respect to the values of R^2, χ^2 and RMSE were evaluated, and the optimal model was selected based on higher values of R^2 and lower values of χ^2 and RMSE. The results of fitting the experimental data with the presented models showed that the best drying model in hot air, infrared and ultrasound dryers were the Page, modified Page, and Handerson and Pabis ones, respectively.

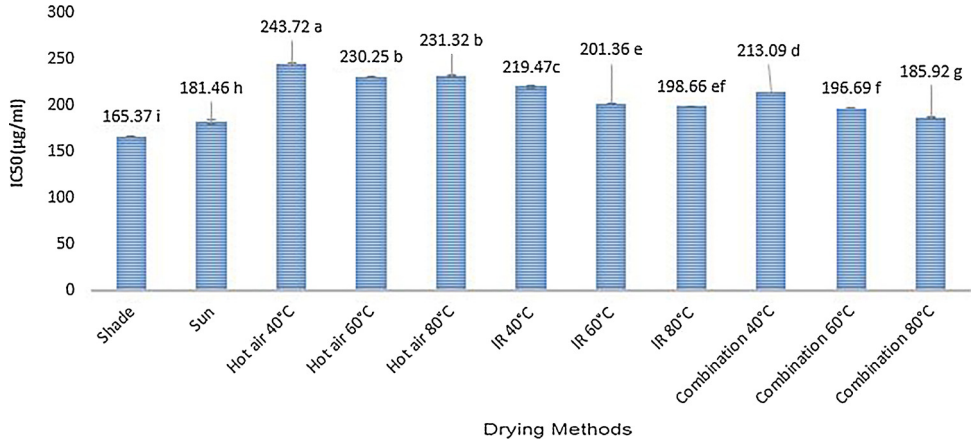

Fig. 5. Comparison of antioxidant activity (IC50) of different drying methods. Different letters showed a significant effect at 5% level.

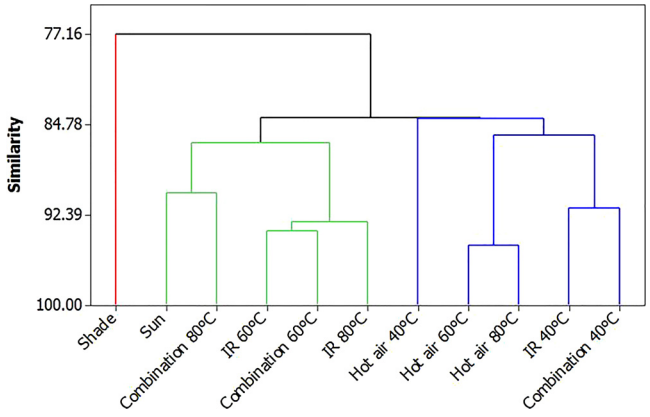

Fig. 6. Dendrogram derived from the clustering analysis of composition (%) of essential oils.

3.3.1. Extracting the hot air drying kinetics model

In the hot air drying, moisture ratio at 40, 60 and 80 °C was obtained and fitted with the existing models (Table 5). Table 6 shows the coefficients and statistical indicators related to these models in the hot air drying method. The Page and modified Page models better than other ones predict moisture ratio based on the temperature of the dryer and the drying time. Values of R^2, χ^2 and RMSE for the two models in the hot air drying method were 0.9988, 0.0087, 0.0094, respectively. Arabhosseini et al. (2009) investigated the drying kinetics of the leaves and the whole plant of Tarragon. The results showed that at the temperature range of 40–90 °C and the air velocity of 0.6 m/s, the Page model was the best mathematical model to describe the drying of Tarragon (Arabhosseini et al., 2009). The results of that study are in agreement with those of the present work.

3.3.2. Extracting the infrared drying kinetics model

Coefficients and statistical indicators related to these models in the infrared method are presented in Table 6. The Page and modified Page predict moisture ratio based on the temperature of the dryer and the drying time, better than other models. Values of R^2, χ^2 and RMSE for the two models in the hot air and infrared methods were 0.9988, 0.0132, 0.0123, respectively. Abe and Afzal (1997) modelled thin layer drying for rice paddy using the infrared drying method, and the Page model was selected as the best model (Abe and Afzal, 1997). The results of these studies are in line with those of the present study.

3.3.3. Extracting the ultrasound-IR-vacuum drying kinetics model

In the ultrasound method, moisture ratio at 40, 50 and 60 °C was obtained and fitted with the existing models. The coefficients and statistical indicators related to these models in the ultrasound method are presented in Table 7. The Handerson and Pabis model at 50 and 60 °C were better than other models to predict moisture ratio based on the temperature of the ultrasound and the drying time. In this method, the values of R^2, χ^2, and RMSE were 0.9986–0.9996, 0.0021–0.0057, and 0.0056–0.0106, respectively.

Kadam et al. (2015) investigated the effect of the ultrasonic pretreatment on the drying kinetics of *Ascophyllum nodosum* using the hot air method by using convective drying (Kadam et al., 2015). In that research, the best laboratory models for the ultrasonic drying were Newton, Midilli, Wang and Singh, and Henderson and Page.

3.3.4. Process optimization and comparison with conventional method

To optimize the process, the target functions are required. These functions were derived using the RSM method. To optimize the process and describe target functions, the boundary conditions were considered according to Table 8.

The boundary conditions such as independent variable (temperature) was considered within the determined variable range. The aim of this optimization was to maximize total phenolic and flavonoid content and antioxidant activity, as well as to minimize the drying time. According to the findings, the proposed optimal temperature points obtained by the software for the hot air, IR, and ultrasonic drying, were 63, 66 and 71 °C, respectively. To investigate the validity of the results, the recommended values were also investigated in experimental conditions. Table 9 shows the values obtained for theoretical and experimental dependent variables in different drying methods.

The other researcher reported that the suggested optimal temperature point for *Satureja bachtiarica* obtained by the software for hot air, infrared and ultrasonic dryer were 54, 53 and 49 °C respectively (Samani et al., 2017). It is worth mentioning that the reason for the difference in optimal temperature between two types of research is due to difference between characteristic of *Satureja bachtiarica* and *Kelussia*. In addition, the optimization in the present research was based on maximizing TFC, TPC and antioxidant activity and minimizing drying time but in other study, the optimization was based on maximizing major essential oil compounds and minimizing of drying time.

4. Conclusion

In the ultrasound-IR-vacuum combination method, when the temperature increased from 40 °C to 80 °C, the TFC increased by 35%. Comparison of the mean values of the treatments in the combination method showed that there was no significant difference between 60 °C and 80° at 5% level. With increasing temperature from 40 °C to 60 °C, the TPC increased by 2.5% and 5%, respectively, in hot air and infrared dying, and then when the temperature reached 80 °C, the TPC in hot air and infrared drying decreased by 4% and 3.5%, respectively. In the combined method, there was a direct correlation between the TPC and the temperature range. With increasing temperature from 40 °C to 60 °C, the TPC increased by 6%. Because that the temperature of 80 °C was lower in for the drying of the *K. odoratissima* sample, the changes in the TFC and TPC were not significant at 60 °C temperature. The highest antioxidant capacity was obtained for dry shade treatment, followed by dry sun treatment and the three temperatures, i.e., 40, 60 and 80 °C, in the combined method. In all three methods, when the temperature increased, antioxidant capacity increased in hot air and infrared drying methods, there was no significant difference between 60 °C and 80 °C temperatures at 5% level, but in the combined method, the antioxidant

Table 5
Results of statistical analyses on the modelling of moisture contents and drying time in the hot air dryer.

Model	40 °C			60 °C			80 °C		
	R^2	χ^2	RMSE	R^2	χ^2	RMSE	R^2	χ^2	RMSE
Newton	0.9885	0.1081	0.033	0.9811	0.1199	0.0373	0.9768	0.1317	0.0416
Page	0.9988	0.0087	0.0094	0.9978	0.0133	0.0125	0.9968	0.0179	0.0155
Modified Page	0.9988	0.0087	0.0094	0.9978	0.0133	0.0125	0.9968	0.0179	0.0155
Henderson and Pabis	0.9936	0.0467	0.0218	0.9905	0.0592	0.0264	0.9874	0.0715	0.0309
Logarithmic	0.9936	0.0467	0.0218	0.9905	0.0592	0.0264	0.9874	0.0715	0.0309
Wang and Singh	0.8865	0.8323	0.0917	0.8782	0.7861	0.5394	0.8698	0.07399	0.987

Table 6
Results of statistical analyses on the modelling of moisture contents and drying time in the Infrared dryer.

Model	40 °C			60 °C			80 °C		
	R^2	χ^2	RMSE	R^2	χ^2	RMSE	R^2	χ^2	RMSE
Newton	0.9809	0.1256	0.0378	0.9689	0.1768	0.0503	0.9848	0.0736	0.0334
Page	0.998	0.0132	0.0123	0.9988	0.0066	0.0098	0.9987	0.0065	0.01
Modified Page	0.991	0.0142	0.0129	0.9911	0.0074	0.0091	0.9982	0.0073	0.02
Henderson and Pabis	0.9907	0.0611	0.0265	0.9871	0.073	0.0325	0.9935	0.0313	0.0219
Logarithmic	0.9907	0.0611	0.0265	0.9871	0.073	0.0325	9935	0.0313	0.0219
Wang and Singh	0.8768	0.8086	0.0959	0.8546	0.8252	0.1086	0.8838	0.5607	0.0922

Table 7
Results of statistical analyses on the modelling of moisture contents and drying time in ultrasound dryer.

Model	40 °C			60 °C			80 °C		
	R^2	χ^2	RMSE	R^2	χ^2	RMSE	R^2	χ^2	RMSE
Newton	0.9854	0.0729	0.0332	0.9763	0.1123	0.429	0.973	0.1076	0.0455
Page	0.9946	0.0268	0.0203	0.9912	0.0417	0.0264	0.9896	0.0416	0.0286
Modified Page	0.9946	0.0268	0.0203	0.9912	0.0417	0.0264	0.9896	0.0416	0.0286
Henderson and Pabis	0.9996	0.0021	0.0056	0.9992	0.0036	0.0077	0.9986	0.0057	0.0106
Logarithmic	0.9946	0.0268	0.0203	0.9912	0.0417	0.0264	0.9896	0.0416	0.0286
Wang and Singh	0.8907	0.5468	0.091	0.8658	o.6358	0.1021	0.8559	0.5751	0.1052

Table 8
Regression relationships between yield, drying time and values of phytochemical compounds.

Parameters	Hot air dryer	Infrared dryer	Ultrasound-IR-vacuum dryer
Total phenol	$25.0328 + 0.5429T - 0.0044T^2$ (R = 0.98) (S = 0.12)	$30.2128 + 0.4264T - 0.0037T^2$ (R = 0.94) (S = 0.24)	$34.9918 + 0.2357T - 0.0017T^2$ (R = 0.95) (S = 0.17)
Total flavonoid	$-14.7702 + 1.0520T - 0.0088T^2$ (R = 0.99) (S = 0.02)	$-21.5383 + 1.3101T - 0.0109T^2$ (R = 0.99) (S = 0.03)	$-11.6425 + 0.8923T - 0.0065T^2$ (R = 0.9849) (S = 0.34)
Antioxidant activity	$335.82 - 3.305T + 0.025T^2$ (R = 0.98) (S = 0.94)	$290.850 - 2.67T + 0.022T^2$ (R = 0.96) (S = 1.01)	$258.73 - 1.519T + 0.012T^2$ (R = 0.93) (S = 0.95)
Drying time	1003.33 − 6.99 T (R = 0.99) (S = 4.08)	978.3 − 7.249 T (R = 0.98) (S = 4.81)	538.3 − 3.25 T (R = 0.99) (S = 4.70)

"T", "R" and "S" Signs indicated temperature (°C), correlation coefficient and standard error, respectively.

Table 9
Amount of theoretical and experimental dependent variables for different drying methods.

	Hot air drying		Infrared drying		ultrasound-IR-vacuum dryer	
	Theoritical	Experimental	Theoritical	Experimental	Theoritical	Experimental
Total phenol	41.7719	41.52	42.2963	42.37	43.1568	43.65
Total flavonoid	16.5786	16.79	17.5657	17.33	18.9443	19.35
Antioxidant activity	226.83	235.41	210.25	210.70	211.373	207.13
Drying time	562.96	550	507.115	515	307.55	315

capacity increased significantly at 5% level among the three temperatures. The study of the similarity of drying methods with respect to the TPC, TFC, and antioxidant capacity showed the least similarity between the shade drying method and other treatments. The results of fitting the experimental data with the presented models showed that the best drying model in hot air, infrared and ultrasound dryers were the Page, modified Page, and Handerson and Pabis, respectively. The proposed optimal temperature points obtained by the response surface method for the hot air, IR, and ultrasonic drying, were 63, 66 and 71 °C, respectively. It should be noted that in the mentioned temperatures, the drying time was reduced to minimum value; however, TFC, TPC and antioxidant activity were increased to their maximum amounts.

Acknowledgements

Research Council of Shahrekord University of Medical Sciences is thankfully acknowledged for the financial support to carrying out the work. Authors are also thankful to Research Council of Shahrekord University to supports provided.

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