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EFFECT OF MODIFICATIONS OF LAVANDIN CONVECTIVE DRYING ON THE COURSE OF THE PROCESS AND ESSENTIAL OIL CONTENT

Summary

The conducted study attempted to link the kinetics of convective drying of lavandin flowers with the quality of the obtained raw material determined by essential oil content, in order to optimize the process. The flowers were dried under natural convection conditions as well as in laboratory dryers with forced drying agent circulation under varying temperatures and periodical time of operation of the heating element in a continuous and intermittent mode. During the drying process, mass changes of the dried material were recorded. Essential oils content was determined using the hydrodistillation method with the use of Clevenger apparatus. For description of the kinetics of water content change, a model resulting from solution of a one-dimensional mass diffusion equation resulting from the Fick's second law under preset initial and boundary conditions was used. A high matching coefficient of the model with the empirically obtained results was determined. Lavandin drying was conducted during the drying rate decrease stage in each of the established drying conditions. The highest amount of essential oils was observed in lavandin flowers dried under intermittent process conditions with initial temperature of 40°C, which was then decreased to 35°C, and with only cyclical provision of heat at 45-minute intervals.
Key words: *lavandin, drying kinetics, essential oils*

WPŁYW MODYFIKACJI SUSZENIA KONWEKCYJNEGO LAWENDYNY NA PRZEBIEG PROCESU I ZAWARTOŚĆ OLEJKÓW ETERYCZNYCH

Streszczenie

W przeprowadzonych badaniach podjęto próbę powiązania kinetyki suszenia konwekcyjnego kwiatów lawendyny z jakością uzyskanego surowca, określoną zawartością olejków eterycznych, celem optymalizacji tego procesu. Kwiaty suszono zarówno w warunkach konwekcji naturalnej, jak i w suszarkach laboratoryjnych z wymuszonym obiegiem czynnika suszącego w zmiennych temperaturach i okresowym czasem pracy elementu grzewczego, w trybie ciągłym i okresowo zmiennym. W czasie procesu suszenia rejestrowano zmiany masy suszonego materiału. Zawartość olejków eterycznych określono metodą hydrodestylacji z wykorzystaniem aparataów Clevengera. Do opisu kinetyki zmian zawartości wody zastosowano model wynikający z rozwiązania jednowymiarowego równania dyfuzji masy wynikającego z II prawa Ficka przy zadanych warunkach początkowych i brzegowych. Stwierdzono wysoki współczynnik jego dopasowania z wynikami uzyskanymi empirycznie. Suszenie lawendyny zachodziło w okresie malejącej szybkości suszenia w każdych, założonych warunkach suszenia. Największą ilość olejków eterycznych odnotowano w kwiatach lawendyny suszonych w okresowo zmiennych warunkach procesowych z początkową temperaturą 40^{o}C, a następnie obniżeniem jej do 35 ^{o}C i tylko cyklicznym dostarczaniu ciepła w odstępach 45 minutowych.
Słowa kluczowe: *lawendyna, kinetyka suszenia, olejki eteryczne*

1. Introduction

The lavender genus is represented by numerous species [1]. A hybrid of the true lavender with the spike lavender, known as lavandin (*Lavandula x intermedia)*, is commonly cultivated around the world and it contains considerably larger amounts of essential oil than the parent forms, with slightly different chemical composition [2, 3], and therefore it has a specific application for cosmetics production [4, 5]. However, study results exist indicating the possibility of its use in the form of natural biocidal products for farming [6] and that it also exhibits antibacterial activity [7]. The industry utilizes mainly the flowers, because they contain several dozen times more oils than the stems [8]. Despite the fact that currently there are numerous methods of plant raw material preservation for storage, the convective drying still appears to be the prevailing one. During the drying process, independently of method and parameters, the material is subjected to changes of physical and chemical and structural characters, sensory characteristics, which influence the quality of the final product [9]. It is possible to enhance the quality of the final product through the proper conducting of this process. The selection of the proper drying method depends on both the type of dried biological material as well as the purpose of the dried material. Thus, the search for perfect methods for different plant species still constitutes a current issue. According to Gunasekaran [10], the course of drying should be evaluated on the basis of different criteria, particularly the rate of the process, energy efficiency, operational costs and quality of the final product.

Essential oils are the most important active bodies in the herbs of the *Labiatae* family used in the pharmaceutical and cosmetics industry. These oils are mixtures of numerous compounds, characterized by high volatility, and thus making the oil raw materials very susceptible to the effect of high temperatures. Thus the paramount importance of selection of the method and drying parameters, particularly temperature. Studies on the drying process of different herbs indicated, that the course of drying kinetics is similar in all of them. It occurs in the period of decreasing drying rate, independently of the assumed method [11, 12, 13, 14]. On the other hand, drying time depends on the selected method and its corresponding parameters, e.g. temperature, microwave power, drying agent flow rate [11, 12, 14]. It was also determined, that the essential oil content is significantly influenced by the method and parameters such as temperature and drying time, whose increase causes a decrease in oil content in the dried material [14, 15]. During low-temperature drying of lavandin it was observed, that high loss of essential oils occurs due to the long duration time of the process. Therefore, attempts were undertaken to limit the phenomenon by proposing a modified method of convective drying.

The objective of the conducted study was to analyze the effect of convective drying conditions (method and temperature) on the kinetics of water content change and the amount of essential oils in lavandin flowers.

2. Study methodology

The study material originated from a university plantation located in northeastern part of Krakow (50.060°N, 19.959°E). Lavandin (*Lavandula intermedia)* cultivar *Grossso* was used in the study. The cultivations were conducted using organic methods on soil with size distribution of heavy loamy sand, mulched with nonwoven crop cover in black color with the substance of $100 g·m^{-2}$. The study was conducted in the fifth year of the plantation use, when the plants originally planted at 45×45 cm spacing already formed a thick lane. The plants were collected in June/July 2015 in full bloom during sunny weather, cutting with pruning scissors whole inflorescences, which were then subjected to drying.

Table. 1. Lavandin inflorescence drying variants
Tab. 1. Warianty suszenia kwiatostanów lawendyny

Drying variant	The course of convective drying process
I	Continuous drying in 40°C
II	Continuous drying in 3540°C
III	Intermittent drying – for 6 hours in 40°C with operation of the fan/ for further 12 h fan only/ then cyclical repetition of the set program 6h heater + fan and 6h fan only
IV	Intermittent drying – for 6 hours in 40°C with operation of the fan/ then cyclical repetition of the set program, 45' fan only, then 45' drying in 35°C + fan
V	Drying under natural conditions in the shade without the air exchange during night.
VI	Natural drying (day and night) in the open, plants exposed to the direct sunlight.

Source: own work / *Źródło*: praca własna

Following the experimental data, which indicated the unfavorable effect of high drying temperatures on the essential oil content in the herbal raw material, low-temperature drying was utilized. Drying was conducted in natural convection in a roofed room, as well as in the sun and laboratory dryers with forced drying agent circulation. In the latter both combinations of variable drying temperatures as well as periodical character of operation of the heating element in continuous as well as intermittent mode were used. Following the literature data [16], intermittent drying was conducted only during the period of decreasing rate of drying. To ensure that this stage was reached in each variant of the experiment, intermittent drying was started first after 6h. In the following part of the work, the drying combinations are described as variants: I, II, III, IV, V, VI. The characteristics of drying process for individual combinations is presented in Table 1.

Sieve load in the dryers was approximately $35.6 g·100 cm^{-2}$. Drying in the dryers was conducted for 3 days, regardless of used combination. On the other hand, in natural conditions drying was conducted until the indication of brittleness and characteristic rustling of the dried material. For each variant of the experiment, 3 dishes containing approximately 5g of flowers each were additionally placed in the dryers in order to record mass change during the drying process. At the initial stage, the measurements of mass change were carried out at 15-20-minute intervals, then with the passage of time and drying duration, the measurement time was extended. The dry mass content in fresh lavandin flowers was determined following the norm PN-R-04013:1988 [17]. On the other hand, the dry matter was calculated on the basis of instantaneous mass values of the dried material and dry weight content determined in the beginning. Water content in lavandin in determined time intervals for individual variants of the drying process was calculated on the basis of changes of mass and dry matter content. The obtained values served for the elaboration of drying curve diagrams and the model of kinetics of water content change.

After termination of the drying process, the flowers were manually separated from the inflorescence axes. For the material obtained in this manner, the mass loss after drying was determined following PN-84/R-87010 [18].

Essential oils content was determined using the hydrodistillation method with the use of Clevenger apparatus. Distillation was conducted for 2 hours, in three repetitions for each combination of the experiment.

The test results were subjected to statistical analysis using the Statistica 12 software. In order to evaluate the significance of essential oil content between the individual drying variants, a single-factor ANOVA was carried out. After the determination of significance of obtained values, homogenous mean groups were separated using the Duncan test. The statistical inference was conducted for the significance level of $P<0.05$.

3. Study results analysis

The kinetics of water content change in time for different drying variants is presented in Figure 1, and the drying rate in Figure 2. For description of the kinetics of water content change, a model resulting from solution of a one-dimensional mass diffusion equation resulting from the Fick's second law under preset initial and boundary conditions was used. The initial conditions were determined using a constant function resulting from the initial water content in the fresh material and the Dirichlett boundary conditions - at the beginning of the process the equilibrium water content is established on the boundary of the area:

- $u = u_0$ for $\tau = 0$ and $0 \le x \le s$,
- $u = u_r$ for $\tau > 0$ and $x = 0$ and $x = s$.

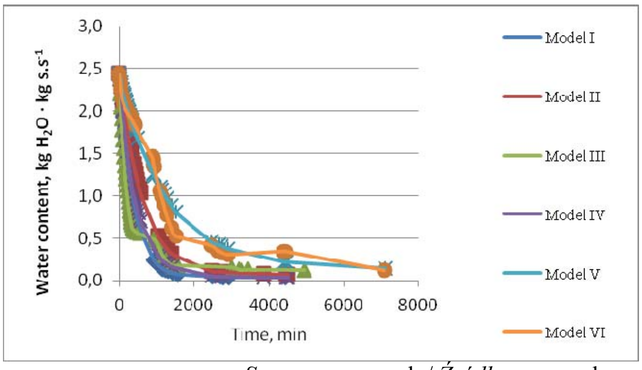
The efficient mass diffusion coefficient D_{ef} was determined using the inverse problem method, on the basis of one series of measurements of water content changes in the studied material. The remaining series were used for the validation of the model.

The course of kinetics of water content changes was described with the following equation:

$$u(\tau) = u_r + \frac{(u_o - u_r)8}{\pi^2}\sum_{n=0}^{\infty} 0{,}1 \exp\left[\backslash\text{-}\backslash 2\backslash{,}\backslash 2\backslash 5\backslash\ \pi^2 \frac{D_{ef}\tau}{s^2}\right], \quad (1)$$

where: D_{ef} – efficient mass diffusion coefficient, $m^2 min^{-1}$,
s – characteristic dimension – thickness of the thin layer, m,
$u(\tau)$ – instantaneous water content, kgH_2O kg d.m.$^{-1}$,
u_o – initial water content, kgH_2O kg d.m.$^{-1}$,
u_r – equilibrium water content, kgH_2O kg d.m.$^{-1}$,
τ – time, min.

The conducted validation of the model confirmed its validity. The calculated local mean square error ranged from 0.0003 to 0.009 kgH_2O kg d.m.$^{-1}$. The determination coefficient for each of the tested variants between the values of water content determined on the basis of measurements and obtained from calculations with the use of the model was $R^2 = 0.99$.

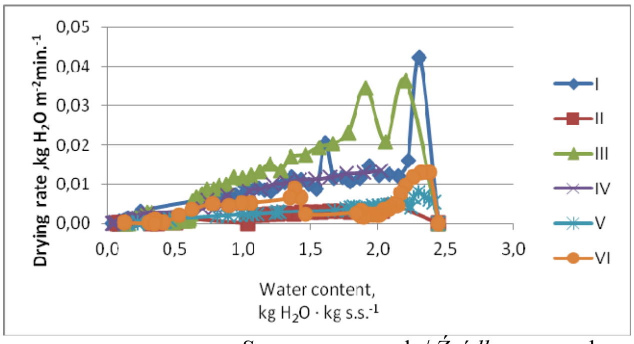


Source: own work / *Źródło*: praca własna

Fig. 1. Drying kinetics curves and model curves

Rys. 1. Krzywe kinetyki suszenia i modelowe

After the assumed drying time, the equilibrium water content depended on the used drying variant – from 0.15 kgH_2Okg d.m.$^{-1}$ for variant V to 0.03 – 0.04 kgH_2Okg d.m.$^{-1}$ for variant I and IV. This difference stems from the conditions of the process. In variant V, lavandin was dried in low temperatures, in a shaded room with gravitational, periodic air exchange, thus air with low ambient temperature as compared to the remaining variants, was moisturized in a short period of time and the partial pressures of steam in the film covering the surface of the material and in ambient air were balanced. On the other hand, in the I and IV variants, the material was dried in a dryer in elevated temperature (40°C), with forced air supply, which considerably sped up water phase transition in the material. As a result of ambient heat supply, the internal energy of the dried material increased, and the continuous forced fresh air supply enabled maintaining pressure and temperature gradients for a longer period of time. Thus, the water transport was and water evaporation from the material were possible. By analyzing the courses of drying rates it can be determined, that lavandin drying occurred during the stage of decrease of the process' rate in each of the used combinations. Similar course of drying kinetics of other herbs dried using other methods: convective – low and high temperatures, microwave, sublimation was found by Wiktor et al. [11], Śledź et al. [12], Rudy et al. [13], Antal et al. [14]. The highest initial drying rate value and its intensive initial decrease were observed in the drying conditions established for variant I – 0.042 $kgH_2Okgd.m.^{-1}$ and for III – 0.036 0.042 $kgH_2Okgd.m.^{-1}$, and the highest in conditions assumed for the variant V (Fig. 2). Such course of drying rate resulted from the conditions in which water evaporated from the material. In the variant I drying was carried out in a constant mode in air heated to 40°C, which was also the case for the first stage of the variant III. Thus, in the initial phase, the process of mass exchange was influenced by two gradients resulting from the difference of: temperatures of the drying agent and the material, and the partial pressures of steam in the ambient drying agent and in the film on the surface of the material. On the other hand, in the case of lavandin dried in the conditions determined for the variant V, the water transport from the material was influenced solely by the gradient of partial pressures of steam over the surface of the material and in the environment.



Source: own work / *Źródło*: praca własna

Fig. 2. Drying rate depending on the water content

Rys. 2. Szybkość suszenia w zależności od zawartości wody

The most stabilized changes of drying rates were observed for the variant IV, for which the highest essential oil content in dried lavandin flowers was also determined, averaging at 7.41 ml·100g d.m.$^{-1}$ (Table 2).

In this case, the heat supply interruptions were established at 45 min. Following the literature data [19, 20, 21; 22], the relaxation period, i.e. interruptions of an intensive moisture removal from the dried material, constitutes a very important element of the intermittent drying. During this period, the humidity and temperature distributions are balanced in the entire mass of the dried material, ensuring their equal distribution. Under such conditions, the water contained in the dried raw material diffuses from deeper layers (which contain its higher content) to the surface. Subsequently, when the heat supply is resumed, the water is evaporated from the surface of the material, which increases drying rate.

The material obtained from the variants I and III is characterized by a similar amount of essential oils. The obtained values were included in the common homogeneous group. The lowest amount of oils were isolated from the raw material dried in the sun, which complies with the experimental data observed for Roman chamomile [23]. Under these conditions the drying rate was related to the variable daily sun exposure. During the night temperature drops and secondary humidification of the dried material occurred.

The raw material dried in natural convection, in the shade, without air exchange during the night, as well as dried in dryer at 35°C, was characterized by similar drying rate and essential oil content. This fact can be explained by similar drying and drying rate curves.

Table 2. Mean essential oil content from individual experiment combinations (ml·100g^{-1} of plant dry matter)
Tab 2. Średnia zawartość olejków eterycznych z poszczególnych kombinacji doświadczenia (ml·100g^{-1}suchej substancji roślinnej)

Drying variant	Essential oil content ml·100g d.m.$^{-1}$ (±SD)
I	6.39±0.11 cd*
II	6.79±0.13 b
III	6.30±0.06 d
IV	7.41±0.13 a
V	6.57±0.23 bc
VI	5.66±0.06 e

*a, b, c, d, e – homogenous groups following the Duncan test
Source: own work / *Źródło*: praca własna

4. Conclusions

1. Lavandin drying was conducted during the stage of drying rate decrease in each of the used variants.
2. The kinetics of the changes of water content during convective drying of lavandin flowers can be described using the model resulting from solution of a one-dimensional equation of mass diffusion formulated on the basis of Fick's second law by determining the initial conditions related to the water content in fresh material and Dirichlet boundary conditions.
3. The use of drying modification under intermittent process conditions with initial temperature of 40°C and then lowering it to 35°C and only periodic provision of heat at 45-minute intervals led to obtaining highest amount of essential oils in the dried material.

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