

MOISTURE DYNAMICS MODELLING IN *HYSSOPI HERBA* DRYING PROCESS BY ACTIVE VENTILATION

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Abstract. Medicinal herbs after their harvesting participate in heat exchange with the environmental, as does each organic and hygroscopic material. Drying helps to conserve the medicinal raw material with the maximal preservation of its quality. The present investigation deals with the influence of the surrounding air factors (temperature and relative humidity) on drying intensity. The study object was aboveground part of hyssop (*Hyssopus officinalis* L.), i.e. *Hyssopi herba* was used as medicinal raw material. The aim of the study was to offer a mathematical model of time-related medicinal herbs moisture dynamics and to show its qualitative agreement with the physical model of diffusion as well as to determine the optimum ventilation intensity of medicinal herbs. Drying of *Hyssopi herba* using active ventilation was investigated. Ventilation intensity and the parameters of the drying agent influenced the processes of moisture dynamics, the total drying time and the quality of *Hyssopi herba*. The basic prerequisites of the drying process have been analysed. A mathematical model of moisture dynamics has been proposed and used to show the dependence of theoretical moisture dynamics on ventilation velocity. The obtained experimental values of moisture content dynamics during the drying of *Hyssopi herba* have been shown to agree with theoretical dependences.

Keywords: Hyssopi herba, moisture content, diffusion, Fick's law, essential oil.

1. Introduction

The quality of medicinal raw material highly depends on the content of active substances. Separate species of medicinal herbs accumulate specific active substances, essential oils being among the most important ones (Bandzaitienė ir kt. 1983; Pourmortazavi and Hajimirsadeghi 2007; Özgüven *et al.* 2007). Evaporation of these volatile substances and the physiological processes taking place in the raw material with high moisture content are decelerated by its proper conservation.

Of all the technological stages of medicinal herb processing, drying consumes up to 60% of all expenses (Müller and Heindl 2006; Мальтри и др. 1979). Drying by ambient air decreases moisture content in the raw. Medicinal herbs and all other plants are hygroscopic, and their drying means evaporating their free moisture (Гинзбург 1973).

During the process, it is important to regulate the parameters such as temperature and relative humidity of the air blown into the medicinal herb layer in order to minimize energy expenditure for ensuring the necessary moisture of dry medicinal herbs and to restrict the evaporation of essential oils. In the paper, the process of active ventilation employing the drying properties of ambient air is discussed.

Medicinal herbs, whose moisture at harvesting reaches 70-80% (Müller and Heindl 2006; Гинзбург 1973), like other hygroscopic organic substances, exchange heat with the surrounding air. Medicinal herbs with a high moisture content in a dry surrounding air undergo drying and evaporate moisture into the ambient air; the process of desorption takes place, whereas dry medicinal herbs in a humid surrounding air absorb moisture from it through the process of absorption (Гинзбург 1973). When absorption equals desorption, i.e. the evaporated moisture flow is equal to the moisture flow absorbed by medicinal herbs, a balance between the medicinal herbs and the environment occurs. The medicinal herbs attain the equilibrium moisture content level corresponding to that of the surrounding air (Kuzmienė ir kt. 1999; Müller and Heindl 2006).

Medicinal herb moisture necessary to ensure their safe preservation for different medicinal herb species varies within 10–13% (Heindl 1999; Müller and Heindl 2006). It depends on air moisture, temperature, the chemical composition of medicinal herbs, the content of different parts of plants (buds, leaves, flowers, fruits, cortex, roots). The optimum moisture content of medicinal herbs is attained when the relative humidity of the drying air is 60–65%. In summer, this level corresponds to the relative humidity of the ambient air in a sunny noon. The drying process is characterized by the drying zone dynamics. It is represented from the beginning of drying, drying zone formation, drying zone shift and drying zone disappearance (Zvicevičius 2003; Мальтри и др. 1979). The drying zone shift with the dry substance, drying raw material and moist raw material is shown in Fig. 1.



Fig. 1. Drying zone dynamics: 1 - dry substance; 2 - dry-ing raw material; 3 - moist raw material (φ in percent is relative air humidity)

The process of medicinal herb drying directly depends on the temperature of the drying air blown into the medicinal herb layer. A one-grade increase of temperature means a 4–5% decrease of relative humidity (Novošinskas ir kt. 1999). In order to preserve the active substances during the medicinal herb drying process, its intensity is highly significant. Also, a proper operation and reliability of the equipment should be ensured.

The aim of the study was to offer a mathematical model of time-related medicinal herb moisture dynamics and to show its qualitative agreement with the physical model of diffusion as well as to determine the optimum ventilation intensity of medicinal herbs.

2. Materials and methods

The study was carried out in 2007 at the Lithuanian University of Agriculture. The study object was the aboveground part of hyssop (*Hyssopus officinalis* L.), i.e. *Hyssopi herba* was used as medicinal raw material.

The following section consists of four parts: the first one discusses the preparation of *Hyssopi herba* for drying by active ventilation and determining essential oils.

The last three parts contribute to the diffusion phenomena, mathematical model, prerequisites, and the equation of moisture balance.

2.1. Drying by active ventilation and determination of essential oils in *Hyssopi herba*

Hyssopi herba was cultivated at the Kaunas Botanical Garden of Vytautas Magnus University. The main active substance is essential oils, their content in the air-dried

Hyssopi herba reaching up to 1% (Ragažinskienė ir kt. 2005). To preserve the active substances, a rather low temperature (30–35 °C) is recommended for drying of *Hyssopi herba* (Adapa and Schoenau 2005; Ragažinskienė ir kt. 2005).

The medicinal raw material was collected during the first flowering. Before drying, it was cut into 4 cm long pieces and dried by active ventilation at the air temperature of the environment. A scheme of the drying circle is presented in Fig. 2.



Fig. 2. Principled scheme of the drying circle: 1 – ventilator; 2 – chamber of constant static pressure; 3 – flexible joint; 4 –valve; 5 – ventilated cylinders filled with medicinal herbs; 6 – temperature and humidity sensors; 7 – secondary ALMEMO measurement unit; 8 – computer

In all the four cylinders (Figs. 2, 5) Hyssopi herba was dried simultaneously at different ventilation intensities: $650 \text{ m}^3/(\text{t-h})$, $2110 \text{ m}^3/(\text{t-h})$, $4310 \text{ m}^3/(\text{t-h})$ and $7330 \text{ m}^3/(\text{t-h})$. The layer height of Hyssopi herba in the cylinders (each one of 0.18 m diameter and 1.2 m height) reached 96 cm, 90 cm, 94 cm and 91 cm, respectively. Ventilation intensity had been regulated by valves before drying (Figs 2, 4). In the process of drying, with the aid of the ALMEMO 3290 (Ahlborn Mess- und Regelungstechnik GmbH, Germany) device equipped with FH A646-21 sensors, the temperature and relative humidity of the drying agent were measured and fixed every 10 minutes (temperature measurement error ± 0.1 °C, relative air humidity error $\pm 2\%$) at the suction hole of the ventilator, in the constant static pressure chamber of the drying rig and in the layers of Hyssopi herba. Initial mass of loaded Hyssopi herba was 5 kg in all the four cylinders. Medicinal herb mass and its average moisture content changes were fixed 2-3 times in 24 hours by weighing a cylinder of the drying rig.

Determination of essential oils in *Hyssopi herba* was performed at Department of Chemistry of Vytautas Magnus University in the Gas Chromatography Laboratory.

The supercritical fluid extraction (SFE) using an HP 7680T SFE module (Hewlett Packard, USA) was performed to prepare samples for gas chromatographic (GC) analysis of essential oils. GC analysis was peformed using HP 5890A gas chromatograph (Hewlett Packard, USA) with a flame ionization detector and DB-5 capillary column ($25 \text{ m} \times 0.33 \text{ mm} \times 0.25 \mu \text{m}$, J&W Scientific, USA). The basic parameters of sample preparation and gas chromatographic analysis are given in Table 1.

Supercritical fluid extraction conditions:	
Amount of dried sample, g	0.5
CO ₂ density, g/ml	0.30
Pressure, bar	91
Extraction time, min	17
Trap column packing	octadecylsilica
Desorption solvent	Heptane
Desorption fraction volume,	
ml	0.7
Gas chromatografphic analysis conditions:	
Injection volume, µl	2
Injector temperature, °C	180
Detector temperature, °C	280
Temperature gradient, °C	from 50 (0) to 190 (2.5),
(temperature variation,	from 190 to 240 (50),
°C/min):	from 240 constant 5 min.

 Table 1. Basic SFE and GC conditions for determination of essential oils

The conditions selected were identic to those used for the sample preparation and analysis of essential oils in *Achillea millefolium* L. (Bimbiraitė *et al.* 2008). For qualitative analysis and determination of relative content of essential oils in percents the maximum content of essential oil determined in a lower layer of the third cylinder (Fig. 2, from ventilator 1) was assumed to be equal to 100%.

2.2. Diffusion

Medicinal herb drying as a physical process means diffusion. Diffusion (lat. *diffusio* – dispersion, dissipation) is a spontaneous dissipation of particles (atoms, molecules) in gases, liquids, solid bodies. Diffusion may involve particles of a substance itself (selfdiffusion), its additive particles or particles of fusing substances because of penetrating into one another.

The measure of diffusion rate is the diffusion coefficient. The highest rate of diffusion occurs in gases, it is slower in liquids and the slowest in solid bodies. In gases, the longer the free flight of molecules, the higher the rate of diffusion.

Diffusion is quantitatively characterized by Fick's law. The amount of a diffused substance is determined by chemical optical, mass spectrometry methods.

Diffusion is highly important in evaporation, condensation, crystal dissolution and crystallization. The spontaneous diffusion of solvent occurs through a semiconductive membrane from a lower-concentration liquid into a liquid of a higher concentration; such a diffusion is of essential significance for living organisms and their metabolic process.

Thus, diffusion is the basic part of numerous chemical, biological and technological processes.

The mathematical modeling of diffusion processes is not quite a novelty in environmental studies. For several years, in our country the air, water and soil pollution has been given rather a comprehensive research. The dynamics of pollution in all these cases is based on diffusion processes. Among the latest works, worth mentioning are investigations of air dustiness in Klaipėda seaport (Baltrėnas *et al.* 2007), mathematical simulation of solid particle dispersion in the air of Vilnius city (Baltrėnas *et al.* 2008). A part of material has been presented in the monographs (Baltrėnas ir Kaulakys 1994; Baltrėnas, Paliulis 2002).

In the most general case, the process of threedimensional diffusion is characterized by the limit problem (Paulauskas 1974):

$$\frac{\partial c}{\partial t} + a \frac{\partial^2 c}{\partial t^2} = D\Delta c - (\mathbf{v}\nabla)c + bc + f(\mathbf{r}, t) - g(\mathbf{r}, t),$$
(1a)

$$c(\mathbf{r},0) = c(\mathbf{r}), \qquad (1b)$$

where $c = c(\mathbf{r}, t)$ is the concentration of a substance undergoing diffusion; \mathbf{r} – coordinate, *m*; *t* – time, *s*; *a* is a constant characterizing the relaxation processes in the substance; *D* is the constant coefficient of diffusion; Δ is the Laplace operator in a chosen system of coordinates; \mathbf{v} is the velocity vector of the environment; ∇ is the Hamiltonian gradient operator; $b = b(\mathbf{r}, t)$ is the external field of constant tension; $f(\mathbf{r}, t)$ and $g(\mathbf{r}, t)$ are the functions of sources and flows, respectively.

2.3. Prerequisites and applications

We shall stress, and it is very important for further exposition, that the classical equation of diffusion in general and in the case of passive transport requires a series of conditions to be imposed (Агеев 2001; Журавлёв 1998):

- the particle size of a diffusing substance is negligibly small and is not a characteristic scale of the model;
- the medium of diffusion is homogeneous and isotropic;
- T = const, i.e. thermodiffusion is absent;
- p = const, i.e. barodiffusion is constant;
- there are no chemical reactions, i.e. sources and outlets of the substance are absent;
- external fields are absent (e.g., there is no electrical diffusion);
- D = const, i.e. at the existing concentration gradients the coefficient of diffusion can be regarded as a constant value;
- the system under study is a two-component one, otherwise changes in the concentration of one of the components at the expense of transport of the other components should be accounted for.

In the cases when at least one of the above conditions is not observed, the mathematical model needs to be adjusted.

The general equation of diffusion and the initial conditions (1a) and (1b) could be simplified. In our case, the process of diffusion is one-dimensional, occurring along a ventilated cylinder: there are no effects of flows and external field, the relaxation processes are negligible, and the function of the source is the function of a point source independent on the coordinate. In most cases, the function of a source is not as simple, and numerical methods are employed for further analysis of the limit problem. After all these presumptions and the consequences following from them we obtain the following limit problem:

$$\frac{\partial c}{\partial t} = D \frac{\partial^2 c}{\partial x^2} - u \frac{\partial c}{\partial x}, \quad c(x,0) = c(x), \quad (2)$$

where *u* is air motion velocity within a bulk of *Hyssopi* herba, m/s; c(x) is the initial concentration of moisture (%).

We shall show that in our case, for the sake of simplicity of the model under analysis, limit problem (2) may be analysed employing not only numerical but also analytical methods. This also allows qualitative conclusions in more complicated cases or using the obtained solutions as the test ones.

First of all we will shift to a moving system of coordinates. To this end, we shall substitute the variables:

$$c(x,t) \rightarrow c(\xi,t), \quad \xi = x - ut.$$
 (3)

After such a substitution of the variables, bordering problem (2) turns into a classical problem of diffusion:

$$\frac{\partial c}{\partial t} = D \frac{\partial^2 c}{\partial x^2}, \quad c(x,0) = c(x).$$
(4)

This allows not only solving equation (4), but also, turning back to the initial variables, to obtain the solution of bordering problem (3):

$$c(x,t) = \frac{Q}{2\sqrt{\pi Dt}} e^{-\frac{(x-ut)^2}{4Dt}},$$
 (5)

where Q is the total moisture content.

2.4. The equation of moisture balance

The medicinal herb moisture is not constant. Nevertheless, we can apply the equation of moisture balance. In the most general case, the distribution of hot air in the layer is described by rather complicated processes of transport. Of all the variables characterizing the process of drying, it is reasonable to single out the flows of mass and energy. From the flow stability expression, we obtain a corresponding equation of balance.

Let the volume V of the air supplied to the ventilation system during the time t contain m_0 of water vapour mass. The medicinal herb mass at the beginning of the drying process is m_1 , at its end $-m_2$, and the air escaping from the ventilation system contains m_0 of water vapour. The mass conservation law tells us that if during the time t the air volume V passes through the system, then $m_0 - m_0 = m_1 - m_2$. Dividing the equation by V, we obtain the equation of concentration:

$$c_0 - c_0 = c_1 - c_2 , (6)$$

where c_0 and c_0 stand for water vapour concentration before and after the process of drying, and c_1 and c_2 are vapour concentrations in the herb before and after drying.

Let the heat quantity change of the hot air flow volume unit before and after the drying process make Δq_0 . The heat quantity needed to evaporate a unit volume mass $m_1 - m_2$ is Δq . The energy conservation law says that

$$\Delta q_0 = \Delta q \ . \tag{7}$$

This equation allows us not only to control the experimental data, but helps us to verify the thermodynamical behavior of the medicinal herb moisture.

3. Results and discussion

3.1. Influence of drying conditions on the quality of *Hyssopi herba* and dependence on airflow

When stored, medicinal raw material in a constant process of heating produces a certain amount of heat which, if not removed, is accumulated by the plants, and the production is overheated. The principle of drying by active ventilation is a cyclic interchange of the two processes – heating and cooling. When medicinal herbs are dried by active ventilation, the drying agent introduces from the surrounding air a certain amount of accumulated heat.

Hyssopi herba was dried at an air temperature of the environment of 22.90 ± 0.12 °C and relative air humidity of $51.30\pm0.32\%$. The drying process lasted until the moisture of *Hyssopi herba* reached the optimum level of 13% (Dachler and Pelzmann 1999; Heindl 1999; Müller and Heindl 2006). Then the airflow to a cylinder was stopped. From 5 kg of the fresh initial mass of *Hyssopi herba* with the moisture content of 72.00±0.36\%, 3.39 kg of water had to be evaporated.

While increasing comparative ventilation intensity (the volume of air getting into the layer per time unit), at first the rate of drying changed insignificantly and became notable only at the airflow velocity of $4310 \text{ m}^3/(\text{t-h})$, *i.e.* when the air filtration rate v reached 0.22 m/s (Fig. 3).



Fig. 3. Average moisture content of *Hyssopi herba* in the process of drying

However, drying by active ventilation with air of the environment at this rate failed to meet the requirement to perform raw drying within 3–5 days (Adapa and Schoenau 2005, Dachler and Pelzmann 1999, Heindl 1999). Even the comparative 7330 m³/(t·h) intensity of ventilation ensured only the limit value of duration: the process of drying lasted 90 hours.

By modelling the process of drying based on solution (5), we have shown that the obtained experimental curves of moisture dynamics (Fig. 3) qualitatively coincide with the theoretical dependences. The adequacy of the employed mathematical model requires statistical substantiation. However, the latter would need a much larger amount of experimental data. In this paper, we will just take advantage of the fact, that, as it is seen in Fig. 3, the functions of fading exponents (5) rather satisfactorily approximate the obtained experimental data.

In the study we used the medicinal raw material that contained essential oils. Changes in moisture and the airflow predetermined their preservation in dried *Hyssopi herba*. Under different ventilation intensity, in the dried *Hyssopi herba* in all the cases the least quantity of volatile substances was lost from the lower layers (Fig. 4).



Fig. 4. Dependence of the relative essential oil content on the comparative air flow in *Hyssopi herba*

3.2. Influence of drying conditions on the quality of *Hyssopi herba* and dependence on drying time

The highest content of essential oils was obtained by employing the air of the environment when the ventilation intensity was 4310 m³/(t·h). This was the result of a rapid drying process of Hyssopi herba, the adequate parameters of the air and their uniform distribution in the medicinal herb mass. From the static pressure chamber, the air with a relative humidity below 65%, while moving up through the lower layer of Hyssopi herba, becomes saturated with moisture, dries the medicinal herbs and loses its sorptive ability. In this case, the damp air that gets in through the upper layers just takes off the heat oozed out by moist herbs and thus prevents their overheating. Hyssopi herba reached this humidity level as soon as 21 hour of ventilation, whereas in the upper layer this process took 72 hours (Fig. 5). Upon the lower layers becoming dry, the formed drying zone from the bottom layer of the herb mass shifts in the direction of the air flow and after a certain period of time reaches the upper layers of the medicinal herb mass.

A rapid drying of the lower layers resulted from a low relative humidity of the air blown into the medicinal herb mass (below 65%) and a comparatively high temperature. In this case, the losses of essential oils were comparatively low (Figs. 4–6). The layer height at the beginning of drying by active ventilation (comparative air flow of 4310 m³/(t·h)) was decreased from 94 cm to 48 cm, and the mass of *Hyssopi herba* – from 5 kg to 1.6 kg, when the average moisture content was reduced up to 13%.



Fig. 5. Airflow relative humidity changes in a bulk of Hyssopi herba (comparative air flow 4310 m³/(t·h))



Fig. 6. Airflow temperature changes in a bulk of Hyssopi herba (comparative air flow 4310 m³/(t·h))

3.3. The layer of medicinal herbs and aerodynamic resistance

The airflow blown into *Hyssopi herba* meets a certain aerodynamic resistance. Aerodynamic resistance is the pressure that should be present in the airflow of a certain intensity to ensure its penetration through the medicinal herb layer. It depends on the thickness and porosity of the layer, pore size and the velocity of the moving air. The aerodynamic resistance to the airflow in a thick layer of ventilated medicinal herbs (static pressure losses Δp) at an air velocity below 0.5 m/s is calculated from the following equation (Weaver *et al.* 1947):

$$\Delta p = k b \rho_{s.m.} v_0^n , \qquad (8)$$

where k is the coefficient of proportionality; b is the layer thickness of the ventilated substance, m; $\rho_{s.m.}$ is the layer density by dry mass, kg/m³; v_0 is the velocity of the air flow in the section of nonfull volume, m/s; n is a power index.

When medicinal herbs are dried with ambient air, the relative energy consumption depends on the comparative airflow; therefore, it is not reasonable to increase the comparative airflow in the absence of positive changes in the quality of dry medicinal herb mass because of considerably higher energy consumption.

4. Conclusions

The moisture of fresh medicinal herbs at harvesting is 70–80%. Their drying by active ventilation to the conditioned moisture level of 10–13% is accompanied by the sorption process. The transport of the diffusing substance depends on the air filtration rate, the difference in partial pressures formed between the drying agent and the bordering layer of particles that wash the product, and the duration of drying.

The moisture balance equation allows not only a quantitative, but also a qualitative determination of residual moisture in medicinal herbs.

The mathematical model of aerodynamic resistance implies that the adequate modelling of porous medium is a fractal set (Miller and Ross 1993).

While modelling the drying process of *Hyssopi herba* applying the diffusion solution, it has been established that the obtained curves of moisture dynamics are in agreement with the results of the physical experiment. The experimental values were adequate to the theoretical dependences.

When *Hyssopi herba* are dried by active ventilation with ambient air, the drying agent transduces the heat accumulated in the air to the drying zone of the medicinal herb layer. Without conditioning the ambient air, it is impossible to avoid temporal variations of the temperature and relative air humidity of the drying agent.

Active ventilation ensures a high quality and safe preservation of *Hyssopi herba* by blowing through the medicinal herb layer a comparative airflow of about 4310 m³/(t·h) with a relative humidity of about 65%. It is not feasible to increase the comparative airflow because of considerably higher energy consumption.

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DRĖGNIO KITIMO MODELIAVIMAS *HYSSOPI HERBA* DŽIOVINANT AKTYVIOSIOS VENTILIACIJOS BŪDU

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Santrauka

Vaistiniai augalai, kaip ir kiekviena organinė higroskopinė medžiaga, dalyvauja šilumos mainų procese su aplinka. Džiovinant siekiama konservuoti vaistinius augalus iki reikiamo drėgnio, kiek įmanoma išsaugant jų kokybę. Nagrinėta aplinkos oro veiksnių (temperatūros ir santykinio oro drėgnio) įtaka džiovinimo intensyvumui. Tyrimams naudota antžeminė vaistinio isopo (*Hyssopus officinalis* L.) dalis, t. y. vaistinė augalinė žaliava – isopų žolė (*Hyssopi herba*). Tyrimų tikslas – sudaryti drėgmės kitimo per tam tikrą laiką matematinį modelį, taikant gautą difuzijos sprendinį, bei nustatyti optimalų ventiliavimo intensyvumą. Tirtas *Hyssopi herba* džiovinimo taikant aktyviąją ventiliaciją procesas. Išanalizavus pagrindines džiovinimo sąlygas, sudarytas drėgmės kitimo priklausomybės nuo ventiliavimo intensyvumo, matematinis modelis. Ventiliavimo intensyvumas ir džiovinimo agento parametrai turėjo įtakos drėgmės mainų procesams, džiovinimo trukmei ir vaistinės augalinės žaliavos kokybei. Gauti drėgnio kitimo džiovinant *Hyssopi herba* rezultatai patvirtino, kad teorinės priklausomybės kokybiškai atitinka tiriamąjį procesą.

Reikšminiai žodžiai: Hyssopi herba, dregnis, difuzija, Fiko desnis, eterinis aliejus.

МОДЕЛИРОВАНИЕ ИЗМЕНЕНИЯ ВЛАЖНОСТИ *НУSSOPI НЕRBA* В ПРОЦЕССЕ СУШКИ С АКТИВНОЙ ВЕНТИЛЯЦИЕЙ

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Резюме

Лекарственные растения, как и всякий органический гигроскопический материал, участвуют в процессе теплообмена с окружающей средой. С помощью сушки требуется консервировать лекарственные растения до нужной влажности, максимально сохраняя их качество. Исследовалось влияние температуры и относительной влажности окружающего воздуха на интенсивность сушки. Объект исследования – наземная часть лекарственного иссопа (*Hyssopus officinalis* L.), т.е. лекарственное сырьё *Hyssopi herba*. Целью исследования было создать математическую модель изменения влажности с течением времени, применяя полученное решение диффузии, и определить оптимальную интенсивность вентиляции. Процесс сушки *Hyssopi herba* исследовался способом активной вентиляции. Различные параметры интенсивности вентиляции и сушильной среды влияли на процесс влагообмена, продолжительность сушки и качество лекарственного сырья. Произведен качественный анализ основных предпосылок процесса сушки. Предложена математическая модель изменения влажности, и на ее основе получена теоретическая зависимость изменения влажности от интенсивности вентиляции. Полученные результаты подтвердили соответствие между теоретической моделью и экспериментальными данными.

Ключевые слова: Hyssopi herba, влажность, диффузия, закон Фика, эфирное масло.

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