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Drying Kinetics and Shrinkage Analysis of *Valeriana Officinalis* Roots

Drying kinetics and shrinkage of valerian plant root (Valeriana officinalis) was investigated during the convective hot air dryer with forced convection mode. Whole root without cutting, root cut into quarters, and root cut into 2 mm thin slices were used in drying experiments. Initial moisture content of roots was 51.2±0.3% and roots were considered to be dry when they lost 68% of the fresh weight and reached the moisture content of 10%. Drying air temperature was set to be 40 and 50°C, air velocity at 1 m/s. The relative humidity of drying air was not controlled and it depended on surroundings. The experimental results were fitted to the five thin layer drying models and according to the non-linear regression analysis Page model was most suitable to describe the drying kinetics. The characteristic drying curves were created for each experimental set and they showed that the samples' preparation strongly influenced the drying process and drying time. Experiments to determine shrinkage of different cell structures of valerian root were carried out for raw material, as well as for dried samples, by using optical and electron microscopy observations and measurements. It was observed that shrinkage processes are significantly dependent of the type of cell tissue and drying air temperature.

Keywords: Valerian roots, convective drying, drying models, material shrinkage, microscopy.

1. INTRODUCTION

The use of medicinal herbs was recognised in many parts of the world before the achievements of modern medicine and the pharmaceutical industry. The root of the valerian plant (*Valeriana officinalis*) is a medicinal herb native to Europe that is widely used for the treatment of tension, irritability, restlessness and insomnia [1, 2]. Postharvest handling practices like handling of roots before drying, removing soil from around valerian roots, selecting drying temperature and technology, and environmental conditions during long-term storage of dried roots, have great effect on the amount of active components in the final product [3].

Many researchers have conducted experimental studies on extraction of essential oil from valerian roots [4, 5]. The level of valerenic acids in commercially available roots from plants was found to average about 3 mg/g dry weight [6]. Valerian can be dried in a wide range of different types of artificial driers. However, studies on the drying behaviour of Valeriana officinalis roots are not present in literature except commercial studies [7, 8]. According to them, the most economical way of drying valerian is in hot air convective drier which comprises a chamber where air heated to a pre-determined temperature. The reduction of drying time of cut roots in a hot air drier with rootlets was estimated to be 20-30% of the time taken by whole roots. The

recommended drying temperature range is 40-70 °C [2].

Several studies have been published on volumetric shrinkage in biological materials. Volume shrinkage in bio-materials depends of moisture content [9]. Heating and loss of water causes stresses in the bio-material cell structure, changes in shape and a decrease in dimensions [10, 11]. Tissue changes usually associated with dehydration are shrunken, shriveled, darkened materials of poor rehydration ability after drying [12]. Water diffusion is directly affected by macro structural and microstructural organization and the collapse of capillary structures and pores could increase the water loss and enhance the shrinkage and collapse of the materials structures [12, 13].

The study on drying kinetics behaviour of Valeriana officinalis roots are rare in the literature. Therefore, the objectives of this study are to determine suitable drying kinetics model that fits own experimental data and to explore the influence of selected drying temperatures on material shrinkage as quality parameter.

2. MATERIALS AND METHODES

2.1 Sample preparation

The roots of valerian (var. *Valeriana officinalis*) were used in the experiments without the separation of roots from the rhizome, Fig.1. Before the experiment, the roots were 30 days stored in the storage at the temperatures of 5 to 15°C. For samples preparation, valerian roots were washed by hand to remove as much soil as possible.

Three sample sizes were used in drying experiments: whole root without cutting, root cut into quarters, and

root cut into 2 mm thin slices. The whole root and root quarters samples were selected for the experiments due to their large presence in valerian industrial production [2, 7]. However, 2 mm thin samples were used in experiments because of the simplicity of sample preparation for microscopic observation [20]. The samples were put in thin-layer on 240x200 mm tray with average net mass of material 0.4 kg and initial moisture content of 51.2±0.3%. The initial moisture content of valerian samples was determined using the oven-drying method [14] with repetition in order to assure accurate initial moisture content average values.

Figure 1. Valeriana officinalis whole root

2.2 Drying experiment

Each experiment involves monitoring and recording the change in net weight of material samples during one invariable drying regime, as in Table 1.

Table 1. Drying experiments

| Experiment no.	Material sample	Drying parameters		
		T, ºC	w, m/s	RH, %
E1	Whole root	40		
E2	Quarters	40		
E3	2 mm slices	40	1	1~4
E4	Whole root	50		
E5	Quarters	50		
E6	2 mm slices	50		

In total, six sets of experiments were conducted, one set for each of the material samples size. Drying air temperature was set to be 40 and 50ºC, and drying air velocity in all experiments was set to be 1 m/s. The selected parameters of the drying process were technological recommended values in valerian industrial production [2, 7]. The relative humidity of drying air was not controlled and it depended on surroundings. The relative humidity of drying air was found to be in the range 1-4%. Valerian roots were considered to be dry when they lost 68% of the fresh weight [2]. The output average moisture content of material was 10%.

2.3 Experimental apparatus

Laboratory scale convective tray dryer were used in experiments, Fig.2. This type of dryer is UOP-8 model like dryer (tray dryer, "Armfield", UK) that is widely used in literature for different medical herbs drying experiments [15-17].

Air heater (*H*) was positioned at the beginning of the tunnel section before the tray section (*TR*) that was placed on digital weight indicator (*DWI*). Fan (*FAN*) was located after the tray section and it provided desired drying air velocity.
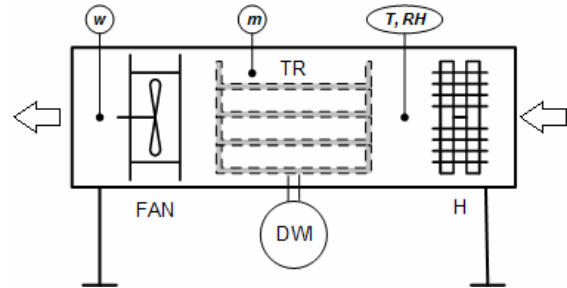
H – Air heater; TR – tray; DWI – digital weight indicator; FAN – Fan; *w* – velocity; *m* – mass; *T* – temperature; *RH* – relative humidity.

Figure 2. Convective laboratory dryer scheme

Measurements of material mass (*m*) and drying air velocity (*w*), temperature (*T*) and relative humidity (*RH*) were performed, Fig.2. The measurement equipment used in experiments have the following accuracies: temperature measurement: ±0.175ºC, relative humidity measurement: ±1%, drying air velocity measurement: ±0.01ms^{-1}, mass measurement: ±0.001kg.

2.4 Drying models

The change of moisture ratio (*MR*) in time was used to describe drying process. The experimental data were fitted to the five well-known thin layer drying models given in Table 2.

Table 2. Mathematical models applied to the characteristic drying curves

Model no	Model name	Model eqation [16, 17]
1	Newton	$MR=exp(-k \cdot \tau)$
2	Page	$MR=exp(-k \cdot \tau^n)$
3	Henderson & Pabis	$MR= a \cdot exp(-k \cdot \tau)$
4	Logaritmic	$MR=a \cdot exp(-k \cdot \tau)+c$
5	Wang & Singh	$MR=1+a \cdot \tau +b \cdot \tau^2$

The moisture ratio was obtained from equation (1)

$$MR = \frac{M - M_e}{M_o - M_e} \qquad (1)$$

where *MR* is the dimensionless moisture ratio, *M*, M_e and M_o are the moisture ratios at any time, initial (at the beginning of the drying process) and equilibrium (at the end of the drying process) moisture content (kg water/kg dry matter) respectively [18].

Non-linear regression analysis was performed for the drying data by using Table Curve 2D (Systat Software Inc. 2002) software [22]. The coefficient of determination (R^2), refer to (2), was the primary criterion for selecting the best model to describe the

drying curves. The higher the values of R^2, the better the goodness of the fit [19].

$$R^2 = 1 - \left[\frac{\sum\limits_{i=1}^{N} \left(MR_{pre.i} - MR_{exp.i} \right)^2}{\sum\limits_{i=1}^{N} \left(\overline{MR}_{pre.i} - MR_{exp.i} \right)^2} \right] \quad (2)$$

where $MR_{exp,i}$ is the i-th experimentally observed moisture ratio, $MR_{pre,i}$ is the i-th predicted moisture ratio and N the number of observations.

2.5 Shrinkage of valerian root

Experiments to determine shrinkage of different cell structures of valerian root were carried out for raw material, as well as for dried samples. The following parameters was monitored on the root cross section, Fig.3:

- area of root (A_k),
- area of root centre cylinder (A_c),
- mean area of cells in exoderm, central mesoderm and inner mesoderm.

The dimension of fresh and dried valerian root sections were measured using optical microscope type Leica DMLS with Leica DC300 digital camera system. The samples were prepared for light microscopy observation by standard paraffin wax method, sectioned by sliding microtome and stained by Alcian blue and Safranin [20].
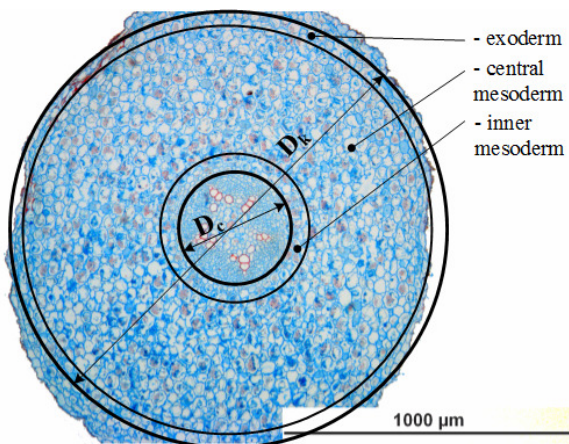

Figure 3. Zones in fresh root sample transversal section in optical microscope (paraffin method, 1000 micron bar is provided on figure)

The dried valerian roots were covered with gold in BALTEC SCD 005 specimen's sputter coater and observed using scanning electron microscope type JEOL JSM-6390, [21].

3. RESULTS AND DISCUSSION

3.1 Drying kinetics

The characteristic drying curves were obtained experimentally for valerian root samples dried at 50°C, Fig.4. Similar results were obtained for the drying temperature of 40°C applied to all three material samples.
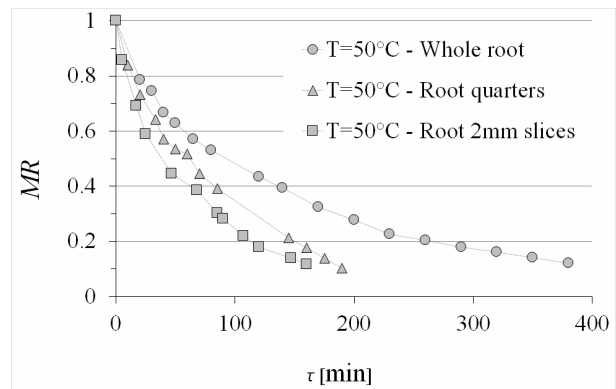

Figure 4. Variations of moisture ratio as a function of time for different material samples dried at 50°C

The results showed that the samples preparation strongly influenced the drying process and drying time. The drying time of whole root was twice longer than drying time of root quarters, regardless the drying temperature. The shortest drying time was gained for the root cut in 2 mm slices. The decrease in material sample size decreases the path distance that moisture travels leaving the inner layers of the material during the drying process. Larger samples require higher drying air temperature for initialization of inner layers water movement.
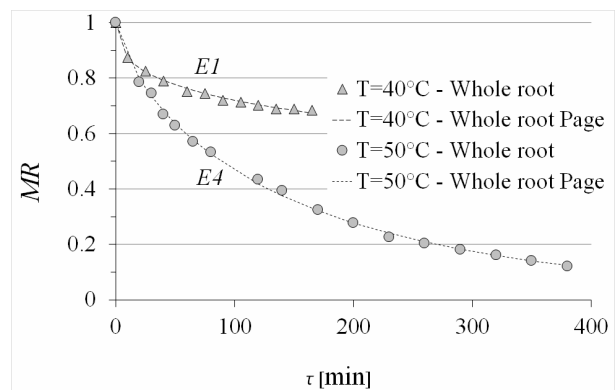

Figure 5. Moisture ratio vs. time for Page model fitting own experimental data for whole root dried at 40 and 50°C

The influence of drying air properties on drying process was compared to the five commonly used literature models. The regression analysis, refer to Table 3, showed that the solutions of Page model provided satisfying match with the results obtained in the experiments $E1$, $E2$, $E4$, $E5$ and $E6$, Fig.5 and Fig.6.
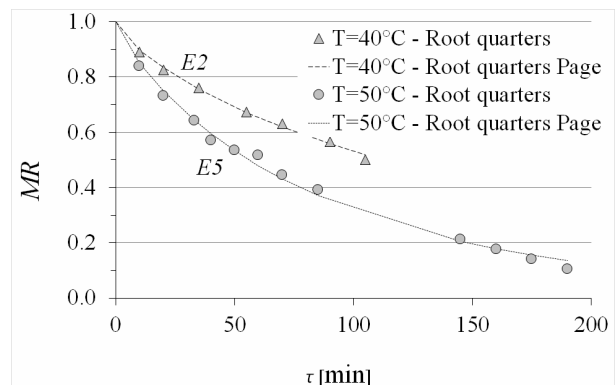

Figure 6. Moisture ratio vs. time for Page model fitting own experimental data for root quarters dried at 40 and 50°C

Table 3. Models statistical analyses results of thin layer tunnel drying of valerian root samples

Model no	Model name	Coefficient of determination for different experimental setups R^2					
		E1	E2	E3	E4	E5	E6
1	Newton	0.50250	0.97188	0.90165	0.95960	0.98570	0.97091
2	Page	**0.99589**	**0.99676**	0.93988	**0.99867**	**0.99520**	**0.99690**
3	Henderson & Pabis	0.81213	0.98660	0.90835	0.98329	0.99186	0.98449
4	Logaritmic	0.97652	0.99169	0.97193	0.99193	0.99242	0.98932
5	Wang & Singh	0.86563	0.98252	**0.97538**	0.93423	0.97084	0.95017

The values of R^2 in Page models were in range 0.99520 to 0.99867. Similar findings were reported for hot air drying of medical herbs [15-18].

In the experiment E3 the best fit was provided with Wang & Singh model, Fig.7. The values of R^2 in Wang & Singh model was 0.97538, and it was slightly higher than 0.93988 of Page model.
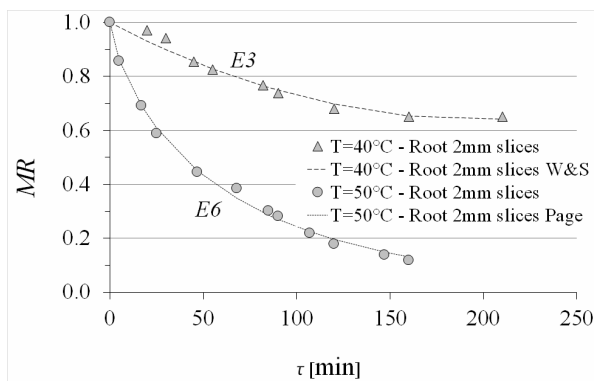

Figure 7. Moisture ratio vs. time for Wang & Singh and Page model fitting own experimental data for root 2 mm slices dried at 40 and 50°C respectively

The change of MR in time was higher at initial stages and then started to decrease with drying time. In all conducted experiments, the decrease in material moisture content was distinct in first 100 minutes. As expected, the increase in drying air temperature will reduce the drying time. Experimental setup E1 has the longest drying time of ~650 minutes, and experimental setup E6 has the shortest drying time of ~200 minutes.

3.2 Shrinkage analysis

Shrinkage of valerian root was determined experimentally by using optical and electron microscopy observations and measurements.
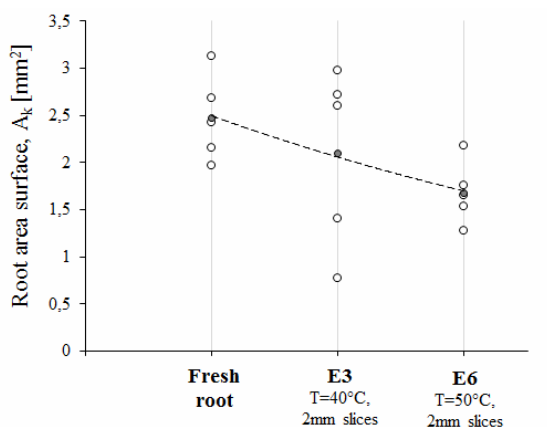

Figure 8. The change of root cross section area regarding the selected drying process

The measured values of root cross-section area and central cylinder, refer to Fig. 8 and Fig. 9, shows that increase in drying temperature will increase the material shrinkage. Initial root cross section dimensions reduced more in higher drying temperatures. The dashed lines, on Fig. 8 and Fig. 9, represents the change of the average values of the area surfaces regarding the selected drying parameters. The average values of measured areas was calculated from several material samples.
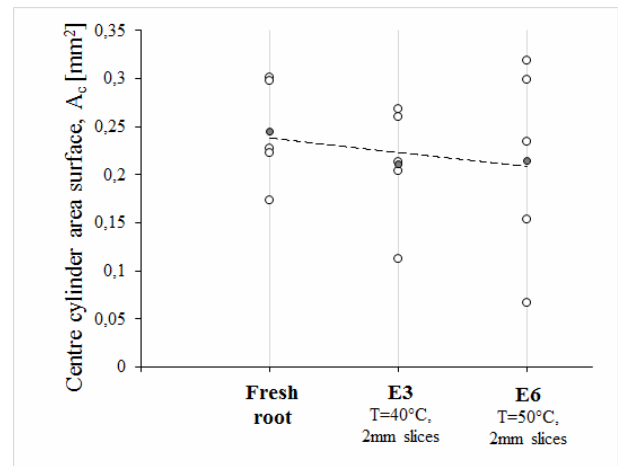

Figure 9. The change of root cross section centre cylinder area regarding the selected drying process
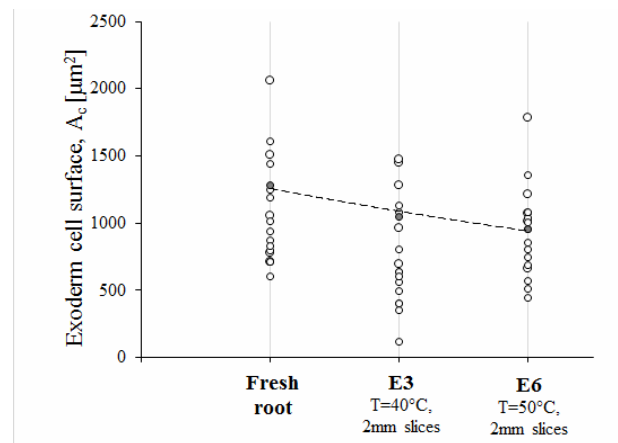

Figure 10. The change of exoderm cell area regarding the selected drying process

The shrinkage of root area was from 2.47 mm2 for fresh sample to 1.68 mm^2, or ~32% reduction. The shrinkage of root center cylinder area was from 0.24 mm^2 for fresh sample to 0.21 mm^2, or ~13% reduction.

The observations of the specific layer cell dimensions in root cross section shows similar results. The average values of the exoderm cell size decreased with increase in drying temperature, Fig.10. The

shrinkage of exoderm cells was from 1280.53 μm² for fresh sample to 958.45 μm², or ~25% reduction.

The similar results were obtained for cell size in central and inner mesoderm. The shrinkage of central mesoderm cells was from 2608.21 μm² for fresh sample to 1639.39 μm², or ~37% reduction, Fig. 11; and for inner mesoderm cells was from 1679.33 μm² for fresh sample to 774.83 μm², or ~54% reduction, Fig. 12.

The results shows that the shrinkage of inside cell layers (inner mesoderm, ~54% reduction) was lager then one from outer layers (exoderm, ~25% reduction). This can be explained with structure of exoderm cell wall, which is richer in suberin.

Plant tissues are made of cells containing more or less amount of intercellular air space. This space tend to collapse when exposed to dehydration. Cell wall is external part of plant cell and its primary role is to keep the shape and structural integrity of the cell. Well-developed secondary cell wall maintains its structure during drying. Cells with thin, primary cell wall, as parenchyma cells, during dehydration change volume and this sort of tissue usually collapsed, refer to Fig. 13 and Fig. 14. The shrinkage processes significantly dependent of type of tissue, i.e. chemical composition of cell walls. Exoderm cells of valerian roots, due to the higher wall suberine content, lose water slower than central than internal mesoderm cells.
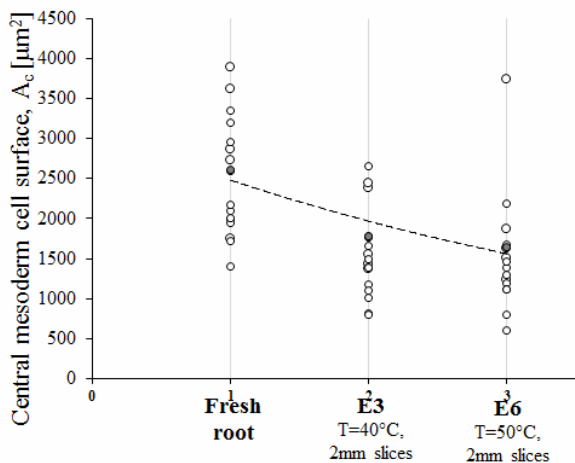

Figure 11. The change of central mesoderm cell area regarding the selected drying process
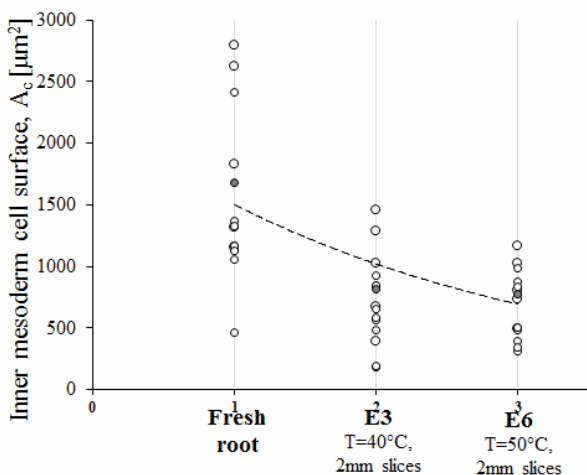

Figure 12. The change of inner mesoderm cell area regarding the selected drying process

Fresh root

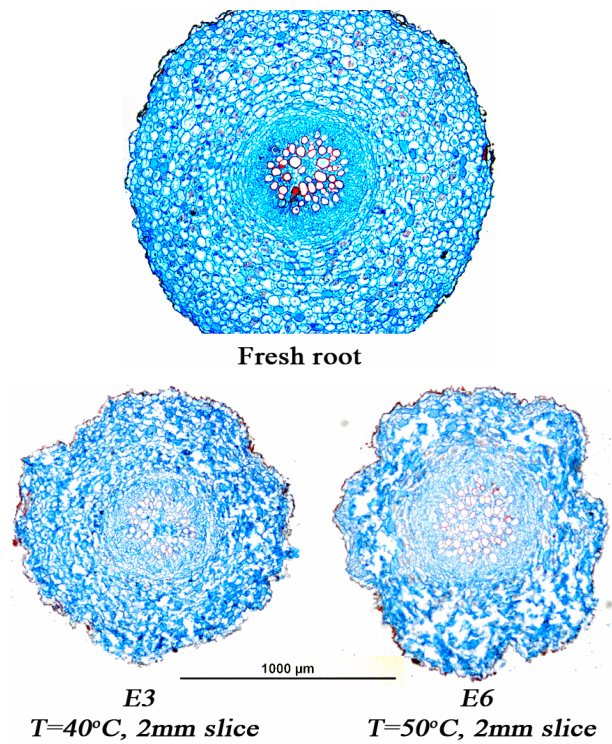E3
T=40ºC, 2mm slice

E6
T=50ºC, 2mm slice

Figure 13. Material shrinkage comparison of dried root samples observed by optical microscopy (paraffin method, 1000 micron bar is provided on figure)

E3,
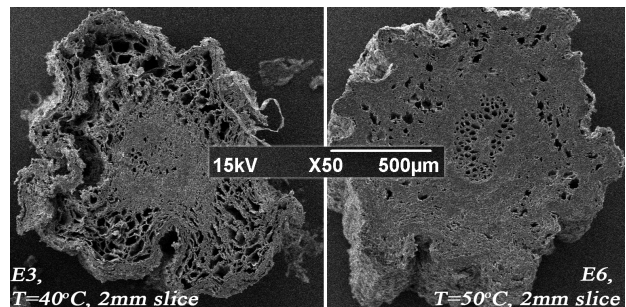*T=40ºC, 2mm slice*

E6,
T=50ºC, 2mm slice

Figure 14. Material shrinkage comparison of dried root samples observed by scanning electron microscope (X50 enlargement, 500 micron bar is provided on figure)

4. CONCLUSION

In this study, the drying kinetics and shrinkage of valerian plant root (*Valeriana officinalis*) was investigated during the convective hot air dryer with forced convection mode.

The conducted study provided the information regarding the most suitable drying kinetics model that fits own experimental data. Also, the influence of selected drying temperatures on material shrinkage as quality parameter was explored and explained for each structural layer of the material.

The drying behaviour was explained using five thin layer drying models. The results showed that the Page model is able to predict the moisture ratio accurately over the period of drying. Since, the coefficients of the Page model are marginally superior, as well as having the highest R^2 values, the model best represents the drying behavior of valerian roots. The values of R^2 in Page models were obtained range 0.99520 to 0.99867.

The shrinkage of valerian root flesh was investigated using optical and electron microscopy observations and

measurements. It was observed that shrinkage processes significantly dependent of type of cell tissue. Results showed that increase in drying temperature will increase the material shrinkage. In all drying experiments, the intercellular air space that tend to collapse when exposed to dehydration. The shrinkage of exoderm cells was slower than shrinkage of central and internal mesodermal cells, which can be explained by the increased content of suberine.

ACKNOWLEDGMENT

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NOMENCLATURE

A	area
a	parameter in model equation, Tab.2
c	parameter in model equation, Tab.2
D	diameter
DWI	digital weight indicator
E_n	experiment number ($n = 1, 2, \dots 6$)
FAN	fan
H	heater
k	parameter in model equation, Tab.2
m	mass

MR	dimensionless moisture ratio
M	moisture ratio
N	number of observations
R^2	coefficient of determination
RH	relative humidity
T	temperature
TR	tray
w	velocity

Greek symbols

τ	time in model equation, Tab.2

Superscripts

c	cylinder
e	equilibrium
exp	experimental
k	root
o	initial
pre	predicted

КИНЕТИКА СУШЕЊА И АНАЛИЗА СКУПЉАЊА БИОМАТЕРИЈАЛА КОРЕЊА VALERIANA OFFICINALIS

И. Златановић, М. Пајић, Д. Ранчић, З. Дајић Стевановић, Д. Дудић

Истраживање кинетике сушења и анализа скупљања биоматеријала корења лековитог биља *Valeriana officinalis* извршено је на конвективној сушари са врућим ваздухом као агенсом сушења. У експериментима су коришћени узорци: цео корен, корен сечен на четвртине и корен сечен на узорке дебљине 2 mm. Почетни садржај влаге у узорцима износио је 51.2±0.3%, а узорак се сматрао сувим онда када садржај влаге опадне на 10%. Температура ваздуха у сушари износила је 40 и 50°C, са брзином струјања од 1 m/s. Релативна влажност ваздуха који доспева на материјал није контролисана и зависила је од спољних услова. Експериментални резултати су анализирани кроз пет најчешће коришћених модела за описивање промене бездимензионог садржаја влаге у времену код сушења у танком слоју, при чему је установљено да се кинетика сушења најбоље описује моделом *Page*-а. Поступцима оптичке и електронске микроскопије извршено је посматрање узорака свежег и осушеног корена при различитим режимима сушења у циљу праћења скупљања материјала. Установљено је да процес скупљања битно зависи од врсте ћелијског ткива у посматраном попречном пресеку материјала и од температурног режима сушења.