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Finite Changes of Bound Water Moisture Content in a Given Volume of Beech Wood

Promjene konačnog sadržaja vezane vode u određenom volumenu drva bukve

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ABSTRACT • *The undesired wood instabilities are connected with the changes of bound water moisture content. The rates of finite changes of bound water moisture content in a given volume of wood were determined in the frame of the specimen dimensions. The derivation is based on the 1st Fick's law and diffusion equation solution in three dimensions. The inverse solution of diffusion equation provided the diffusion coefficients in the principal anatomical directions. Beech wood was tested. The nonlinear regression is involved in the inverse solution. Therefore, the starting values of diffusion coefficients were computed according to the proposed three dimensional square root scheme. The evaluation of data revealed the random character of transport characteristics. The sufficient condition for the slowest finite change of bound water moisture content in a given volume of beech wood is the ratio 8.7:1.5:1.0 of dimensions in longitudinal, radial and tangential directions.*

Key words: *beech wood, diffusion coefficient, moisture content*

SAŽETAK • *Nepoželjna pojava nestabilnosti drva povezana je s promjenom sadržaja vode vezane u drvu. Promjene konačnog sadržaja vode vezane u određenom volumenu drva određene su u ovisno o dimenzijama uzorka. Derivacija se temelji na 1. Fickovu zakonu i rješenju jednadžbe difuzije u tri dimenzije. Inverzna rješenja jednadžbe difuzije daju koeficijente difuzije u glavnim anatomskim smjerovima. Test je proveden na uzorcima bukovine. Nelinearna je regresija uključena u inverzna rješenja, stoga su početne vrijednosti koeficijenata difuzije izračunane prema predloženoj shemi trodimenzionalnoga kvadratnog korijena. Ocjena rezultata otkrila je svojstvo slučajnosti kretanja vode. Dovoljan uvjet za najsporiju promjenu konačnog sadržaja vode vezane u određenom volumenu bukovine omjer je dimenzija 8,7 : 1,5 : 1,0 u uzdužnome, radijalnome i tangencijalnom smjeru.*

Ključne riječi: *bukovina, difuzijski koeficijent, sadržaj vode*

1 INTRODUCTION
1. UVOD

The wood products should be used in an environment that can ensure constant moisture content. Otherwise, the undesired mass and dimensional changes can occur. It is hard to maintain the constant moisture content for a long wood product life cycle; this proved inevitable in most observations and experiments. Wood equilibrates the bound water moisture content when it is in contact with humid air. The humid air should have constant parameters, namely relative humidity and

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temperature (Langmuir, 1918; Brunauer *et al*., 1938; Dent, 1977). From the view point of drying, there is a lot of humid air with given parameters inside a drying kiln or inside a climatic chamber. The change of the equilibrium moisture content is accompanied with the change of the humid air parameters. The change can be infinitesimal or finite. The infinitesimal changes are connected with the phenomenon of the sorption isotherm. The finite changes only deal with the diffusion of the bound water as far as humid air has never been saturated and free water has not been present in wood. Then bound water mass flux $\vec{q}$ is induced, which is proportional to the concentration gradient as the 1st Fick's law states:

$$\vec{q} = -\overline{\overline{D}}\, grad(c) \qquad (1)$$

Where c is concentration defined as the water mass in wood volume, D is diffusion coefficient as the second order tensor.

Diffusion equation describes the finite changes of the bound water moisture content and introduces the time of the diffusion in its solution. Also solutions introduce the diffusion coefficient D and methods of its measurement can be arranged when the particular solutions of diffusion equation are known (Hrčka and Babiak, 1999):

1. zero time derivative of concentration – steady diffusion – King's method (Skaar, 1954), cup methods (Choong, 1965; Siau, 1995)
2. nonzero time derivative of concentration – unsteady diffusion
   1. variable initial condition – moment method (Cunningham *et al*., 1989)
   2. constant initial condition – sorption method

I. kind boundary condition (Dirichlet condition)

1. constant boundary concentration – solution from Fourier series (log scheme, half-time scheme (Skaar, 1954; Chen *et al*., 1994)); solution from Laplace transform (square root scheme (Comstock, 1963; Stamm, 1964))
2. variable boundary concentration – relaxation time method (Babiak et al., 1989)

II. kind boundary condition (Newman condition) – average concentration or flux method (Babiak *et al*., 1989)

III. kind boundary condition (Robin condition) – including mass transfer coefficient - half-time method (Alpatkina, 1968; Choong and Skaar, 1969; Choong and Skaar, 1972; Liu, 1989); other approaching methods (Söderström and Salin, 1993; Dincer and Dost, 1996).

There are some methods for analyzing the constancy of diffusion coefficient – Egner's method (Skaar, 1954), Rosenkilde and Arfvidsson (1997), and methods based on the wood drying data (Dekrét and Trebula, 1999) if moisture field is known, or numerical methods for evaluating the sorption measurements if the dependency of diffusion coefficient on moisture content is prescribed (Olek *et al*., 2005). Siau (1984) published the exponential function of the diffusion coefficient on moisture content and Stamm (1964) and Choong (1963) found out its increasing dependence on temperature according to Arhenius equation. Also Kang and Hart (1997) confirmed such relationship experimentally. Olek *et al*. (2011) showed that the application of the diffusion coefficient function on water content improved only insignificantly the prediction of change of the average bound water moisture content in time and the process was modeled by modified convective boundary condition. Yeo and Smith (2005) proved the existence of convective mass transfer coefficient based on the experiment. The infinite slab hypothesis was defeated by Droin-Josserand *et al*. (1989), who investigated the process of moisture adsorption in three dimensions by wood samples of cubic and parallelepiped shapes. Baronas *et al*. (2001) guarantee a better prediction with a two-dimensional model than with a one-dimensional model in the case of extremely long specimen if the ratio of the width to thickness of the specimen is less than 10; otherwise some edges should be heavily coated. The computational disadvantages of three dimensional models in comparison with two dimensional models are reported by Da Silva *et al*. (2014).

The water flux enters wood volume modeled as continuum, through the wood surface. A given volume can have different surface areas. Therefore, there is a lot of ways how to equilibrate the finite change of the wood equilibrium moisture content of a given volume. The hypothesis of this research is the existence of an extreme. Such extreme cannot depend on the duration of the process. The aim of the contribution is the determination of the extreme, non-zero finite equilibrium moisture content change in a given wood volume. The second hypothesis is that if the volume of cube is enveloped by the smallest surface, than the extreme will be minimal.

2 METHOD AND MATERIAL
2. METODA I MATERIJAL

The finite change of moisture content in a given volume of wood can be found using solution of diffusion equation in the form:

$$D_1 \frac{\partial^2 c}{\partial x_1^2} + D_2 \frac{\partial^2 c}{\partial x_2^2} + D_3 \frac{\partial^2 c}{\partial x_3^2} = \frac{\partial c}{\partial t} \qquad (2)$$

Where i=1, 2, 3; xi are spatial coordinates of the range $\langle -L_i, L_i \rangle$, Di is diffusion coefficient in the i^{th} principal anatomical direction (1–longitudinal, 2-radial, 3-tangential), t is time.

The boundary and initial conditions are of the form:

$$\left(-\mathrm{L_i}\right)\left.\frac{\partial \mathrm{c}}{\partial \mathrm{x_i}}\right|_{\mathrm{x_i=L_i}} = \mathrm{Bi}\left(\left.\mathrm{c}\right|_{\mathrm{x_i=L_i}} - \mathrm{c}_\infty\right) \qquad (3)$$

$$\left.\frac{\partial \mathrm{c}}{\partial \mathrm{x_i}}\right|_{\mathrm{x_i=0}} = 0 \qquad (4)$$

$$c(x,0) = c_0 \qquad (5)$$

Where Bi is Biot number; c_∞ is equilibrium concentration; c_0 is the initial, constant concentration.

The particular solution is found in the form:

$$\frac{c-c_\infty}{c_0-c_\infty}=\sum_{n=1}^{\infty}\prod_{i=1}^{3}A_{in}cos\left(\mu_{in}\frac{x_i}{L_i}\right)e^{-\left(\mu_{1n}^2\frac{D_1}{L_1^2}+\mu_{2n}^2\frac{D_2}{L_2^2}+\mu_{3n}^2\frac{D_3}{L_3^2}\right)t} \quad (6)$$

Where $A_{in}=\frac{2sin(\mu_{in})}{\mu_{in}+sin(\mu_{in})cos(\mu_{in})}$, $\mu_n tg(\mu_n)=Bi_i$ are the roots of characteristic equation and other quantities as previously defined.

A similar solution can be found for heat transfer in the study of Luikov (1968). The moisture content w of the parallelepiped is proportional to the integral of concentration over the wood volume:

$$w=\frac{8}{m_0}\iiint_{000}^{L_1\,L_2L_3}c\,dx\,dy\,dz \quad (7)$$

Where m_0 is the specimen oven dry mass.

The model (6) contains six unknown parameters (three diffusion coefficients and three Biot numbers). It is, therefore, desired to use three specimens of different dimensions as far as the function of moisture content in time is used for their estimation. Then, the least square method and nonlinear regression techniques can be applied. Therefore, starting values for six unknown parameters must be provided. The three dimensional square root scheme was developed to find the staring values of diffusion coefficients.

$$\begin{pmatrix}\frac{1}{L_{11}} & \frac{1}{L_{21}} & \frac{1}{L_{31}}\\ \frac{1}{L_{12}} & \frac{1}{L_{22}} & \frac{1}{L_{32}}\\ \frac{1}{L_{13}} & \frac{1}{L_{23}} & \frac{1}{L_{33}}\end{pmatrix}^{-1}\begin{pmatrix}\frac{\sqrt{\pi}}{2\sqrt{t}}\frac{w_1}{w_{\infty1}}\\ \frac{\sqrt{\pi}}{2\sqrt{t}}\frac{w_2}{w_{\infty2}}\\ \frac{\sqrt{\pi}}{2\sqrt{t}}\frac{w_3}{w_{\infty3}}\end{pmatrix}=\begin{pmatrix}\sqrt{D_1}\\ \sqrt{D_2}\\ \sqrt{D_3}\end{pmatrix} \quad (8)$$

where wi, $wi\infty$ are values of the i^{th} specimen's moisture content at time t and at equilibrium, Lik are the i^{th} specimen's half of the total dimension in the k^{th} anatomical direction, i, k=1, 2, 3. Initial values of the Biot number were set to the value of 10 on the basis of previous analysis and experiments (Babiak, 1996; Hrčka, 2008). The precision of moisture content estimation was evaluated according to the residual standard deviation σres:

$$\sigma_{res}=\sqrt{\frac{\sum_{j=1}^{N}\left(w_{j\,theoretical}-w_{j\,experimental}\right)^2}{N-K}} \quad (9)$$

Where K is the number of estimated parameters (K=6).

Beech wood (*Fagus sylvatica, L.*) was selected as the experimental material. The specimens were free of visible defects. Beech is the most abundant and harvested tree species (32.7 % of total trees) in Slovakia (Green report, 2014). The experiment was conducted in the climatic chamber with controlled temperature and relative humidity. Mass was determined on the same balances, with the use of tare before every mass measurement. The mass was recorded to 1 mg. The dimensions were measured with calliper in millimetres to two decimal places. The beech specimens with dimensions of 5, 10, 15, 20, 25, 30 mm (denoted as A, B, C, D, E, F) in radial and tangential directions and 150 mm in longitudinal direction were used as experimental material. The specimens were cut by sharp circular saw from radial board initially located in the outer part of the stem. Six replicates represented every dimension. Zero moisture content was the initial condition and the experiment was stopped when the mass of every specimen started to slightly oscillate in the humid air at the temperature of 20±0.4 °C and relative humidity of 85±3 %.

3 RESULTS AND DISCUSSION
3. REZULTATI I RASPRAVA

The diffusion coefficient and Biot number, evaluated from adsorption experiment of beech wood in humid air are random variables (Comstock, 1963; Dekrét and Kurjatko, 1986; Hrčka, 2008; Sonderegger *et al.*, 2011). The critical p-level value was set to 5 % as a variability of data can be assumed. Even though the parameters are only positive numbers, they fulfil the requirement of the normal distribution. The volume was the factor in ANOVA with 5 degrees of freedom. The Cochran test proved non-significant differences between variances and one-way ANOVA, 7-times repeated, proved non-significant influence of volume on transport characteristic values. The residual standard deviation was the only exception, and Duncan's test marked the value of A group significantly different (*p*-level 0.04).

The anatomical direction is the significant factor for the diffusion coefficient. It has the largest average value in longitudinal direction and the smallest value in tangential one. The null hypothesis (H0) for Biot numbers cannot be refused on the bases of experimental data, the averages differed on the fifth decimal place, and therefore unique average value is assumed for it. Table 1 shows statistical data of bound water diffusion characteristics of beech wood obtained on the basis of the experiment.

Additional results are also shown in Figure 1.

The residual standard deviation reached the value of 0.194 % and its individual values for different volumes are embedded in Table 2.

The change in moisture content is proportional to fluxes through the surfaces. The areas of surfaces are

Table 1 Statistical data of bound water diffusion characteristics of beech wood obtained by the experiment (1–longitudinal, 2-radial, 3-tangential)
Tablica 1. Statistički podaci obilježja difuzije vode vezane u uzorcima bukovine dobiveni eksperimentom (1 – longitudinalni smjer, 2 – radijalni smjer, 3 – tangencijalni smjer)

	$D_1\cdot10^{-12}$ $m^2\cdot s^{-1}$	$D_2\cdot10^{-12}$ $m^2\cdot s^{-1}$	$D_3\cdot10^{-12}$ $m^2\cdot s^{-1}$	**Bi**
Average *Prosječno*	1510	45.3	20.0	20.9
Stand. deviation *Standardna devijacija*	1.9	8.1	6.3	0.5
No. of observations *Broj mjerenja*	60	60	60	60

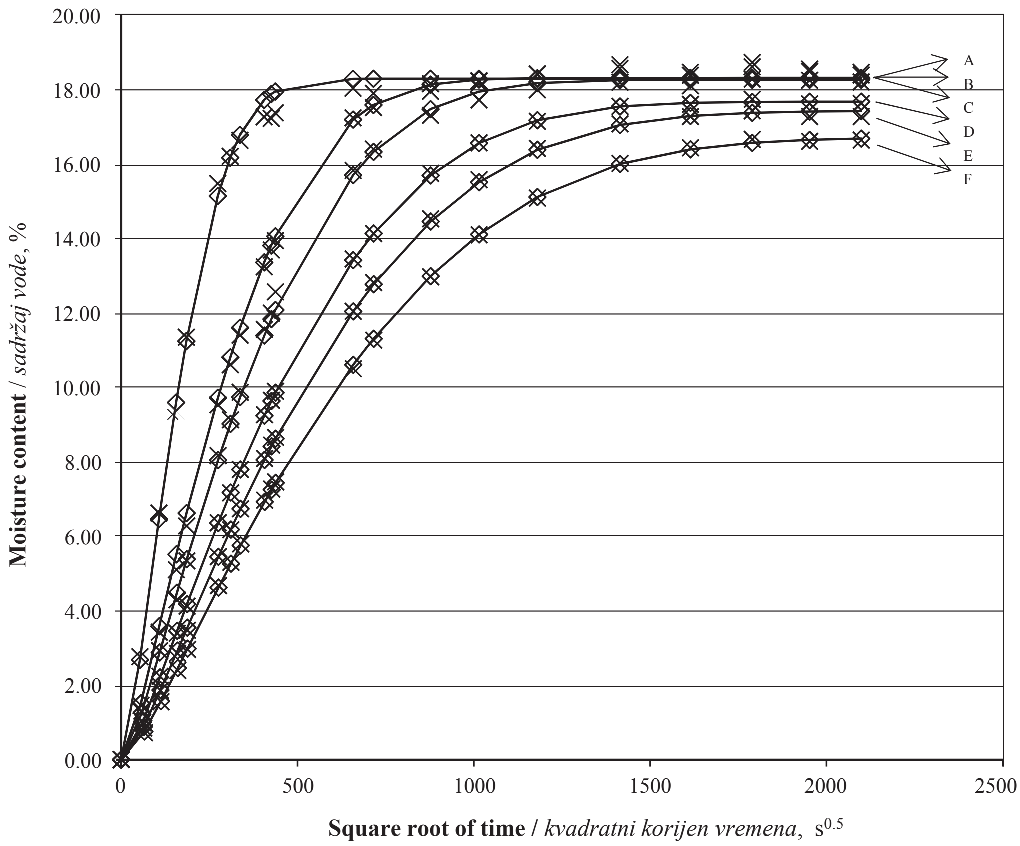

Figure 1 Graphs of bound water moisture content as function of square root of time for different beech wood cross sections, the experimental data are marked as crosses (thicknesses: A – 5 mm, B – 10 mm, C – 15 mm, D – 20 mm, E – 25 mm, F – 30 mm)
Slika 1. Prikaz funkcije sadržaja vezane vode u ovisnosti o drugom korijenu vremena za različite poprečne presjeke bukova drva (debljine: A – 5 mm, B – 10 mm, C – 15 mm, D – 20 mm, E – 25 mm, F – 30 mm)

the proportionality factor for such change. The volume can be enclosed with different surface areas. The extreme values of areas must fulfil the following conditions:

$$\frac{\partial^2 w}{\partial L_j \partial t} = 0 \tag{10}$$

Where L_j denotes the dimension in the j^{th} anatomical direction (for example in radial, and then in tangential).

Then the following relationship is valid for linear orthotropic material such as radial board (and also for cylindrical orthotropic material such as wood):

$$\frac{D_1}{L_1^2} = \frac{D_2}{L_2^2} = \frac{D_3}{L_3^2} \tag{11}$$

Where L_1, L_2 and L_3 are half of the total dimensions of the wood specimen in longitudinal, radial and tangential directions. The determinant ($i \neq j$):

$$\begin{vmatrix} \frac{\partial^3 w}{\partial L_i^2 \partial t} & \frac{\partial^3 w}{\partial L_i \partial L_j \partial t} \\ \frac{\partial^3 w}{\partial L_j \partial L_i \partial t} & \frac{\partial^3 w}{\partial L_j^2 \partial t} \end{vmatrix} \tag{12}$$

is positive and extremes will occur. If sorption occurs as diffusion, the first term is negative; then the conditions (11) are sufficient to characterize the local minimum of the moisture content change in time during bound water diffusion in wood. If desorption occurs as diffusion, then the gradient is opposite to the outside normal of the surface and its absolute value must be involved in determinant (12) instead of the actual value. Also, the local minimum occurs. Once the wood volume is set and diffusion coefficients for wood are determined, the dimensions are determined for the slowest finite change of bound water moisture content.

Table 2 Values of residual standard deviations for different volumes (thicknesses: A – 5 mm, B – 10 mm, C – 15 mm, D – 20 mm, E – 25 mm, F – 30 mm)
Tablica 2. Vrijednosti rezidualne standardne devijacije za različite volumene (A – 5 mm, B – 10 mm, C – 15 mm, D – 20 mm, E – 25 mm, F – 30 mm)

Volume / *Volumen*	**A**	**B**	**C**	**D**	**E**	**F**	**Average** *Prosjek*
Res. stand. deviation, % *Rezidualna standardna devijacija, %*	0.456	0.195	0.199	0.097	0.104	0.117	0.194

The extreme values of dimensions can be computed as follows:

$$L_1^3 = \frac{V}{8}\frac{D_1}{\sqrt{D_2 D_3}} \quad (13)$$

$$L_2^3 = \frac{V}{8}\frac{D_2}{\sqrt{D_1 D_3}} \quad (14)$$

$$L_3^3 = \frac{V}{8}\frac{D_3}{\sqrt{D_2 D_1}} \quad (15)$$

Where V is a given wood volume.

The optimal beech wood dimensions of unit volume for the finite change of bound water moisture content fulfil the ratio (longitudinal : radial : tangential):

8.7 : 1.5 : 1.0

Then, due to the finite change of bound water equilibrium moisture content at a given Biot number of 20.9 and temperature of 20 °C, the diffusion exhibits the slowest process in beech wood.

4 CONCLUSIONS
4. ZAKLJUČAK

There were a lot of ways to equilibrate the finite change in the wood equilibrium moisture content of a given volume. The extreme rate of finite change of bound water moisture content in a given volume of wood was found and the hypothesis of this research was confirmed. Such extreme does not depend on the duration of the process. The extreme is unique and minimal. The minimum for a given volume depends solely on diffusion coefficient eigenvalues in the principal anatomical directions. The diffusion coefficient is a random variable and, therefore, the whole procedure can be repeated and the results are reproducible.

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