Diffusion Coefficient and Equilibrium Moisture Content of Different Wood Species Degraded with *Trametes versicolor*

Barbora Slováčková,[a,*] Jarmila Schmidtová,[b] Richard Hrčka,[a] and Oľga Mišíková [a]

The degradation of wood changes various properties; these changes can favor its usage in particular instances, *e.g*., as an insulation material. Knowledge of the moisture content and movement of moisture in building materials is crucial. The primary focus of this paper is the diffusion coefficient and equilibrium moisture content of three wood species after degradation *via Trametes versicolor*. Values for the diffusion coefficients were determined under different conditions: a temperature of 20 °C ± 2 °C; and 3 relative air-humidity settings, *i.e.*, 30% ± 3%, 60% ± 3%, and 96% ± 3%. The differences in the longitudinal and transversal directions were statistically significant for all conditions, while the differences in the diffusion coefficients were non-significant for the first two relative-air-humidity settings. A portion of the diffusion coefficient calculation data was used to develop a sorption isotherm for all wood species. The equilibrium moisture content of the degraded wood was determined for each condition. Duncan's multiple-range test showed that the wood species was a significant factor; therefore, the isotherm had to be plotted for each wood species. The number of sorption sites in the monolayer in degraded spruce wood was different from the number in degraded beech and oak wood.

Keywords: Diffusion coefficient; Equilibrium moisture content; Trametes versicolor; Spruce; Beech; Oak heartwood; Sorption

*Contact information: a: Department of Wood Science, Technical University in Zvolen, T. G. Masaryka 24, Zvolen 96001 Slovakia; b: Department of Mathematics and Descriptive Geometry, Technical University in Zvolen, T. G. Masaryka 24, Zvolen 96001 Slovakia; *Corresponding author: xslovackova@is.tuzvo.sk*

INTRODUCTION

Thermal insulation products based on renewable resources are emerging on the market as an alternative to the usual thermal insulation based on non-renewable resources. Using a bio-based thermal insulation has multiple advantages; the primary advantage is the raw materials used to produce bio-based thermal insulation are either renewable resources or by-products of agricultural output. In addition, the other ingredients used in bio-based thermal insulation are harmless to the environment.

One of the most important potential advantages is the fact that natural raw materials are highly hygroscopic. Hygroscopic materials have the capacity of adsorbing and desorbing water vapor, which contributes to moderating the extreme variation in humidity found in indoor environments (Simonson *et al.* 2004c; Osanyintola and Simonson 2006; Qin *et al.* 2011; Palumbo *et al.* 2016). Several researchers (Simonson *et al.* 2001, 2004a,b,c; Rode *et al.* 2002; Holm *et al.* 2004; Peukhuri *et al.* 2004; Salonvaara *et al.* 2004; Svennberg *et al.* 2004; Hameury 2005) have studied the use of various hygroscopic materials to moderate indoor humidity levels. The studies have shown that hygroscopic

materials are able to moderate the indoor humidity levels and thus improve the thermal comfort and perceived air quality in buildings (Hameury 2005; Simonson 2005).

An experiment on the dynamic vapor sorption of bio-based insulations and expanded polystyrene by Palumbo *et al.* (2018) showed that the natural materials started to absorb water vapor immediately. The researched bio-based materials were made from wood wool and corn pith. At a relative air humidity of 90%, the wood wool reached a moisture content of 15% and the corn pith reached a moisture content of 35%. The expanded polystyrene did not absorb water until it reached a point when the relative air humidity was high enough (approximately 80%) (Palumbo *et al.* 2018).

Wood is also a hygroscopic material, and the moisture-related properties in wood affect its other properties. It is a well-known fact that the moisture content of wood strongly depends on the conditions of its surroundings, *e.g.*, the air temperature, relative air humidity, and other factors (Siau 1995). According to the properties of wood and the conditions of its environment, either sorption or desorption occurs. The processes of sorption and desorption can be expressed with a diffusion-coefficient and a sorption-isotherm model (Požgaj *et al.* 1997). On the basis of these properties and models, it is possible to predict the moisture behavior of wood in different conditions.

Diffusion in a material is mass transfer and in wood, it describes the movement of bound water due to a difference in concentration. Diffusion is mentioned in various wood-manufacturing processes, *e.g.*, wood drying, the movement of moisture through the layers of an exterior wall or interior furniture, or the reaction of wood to changes in the relative moisture in the manufacturing process (Siau 1995). Diffusion is described by Fick's first and second laws. The integral form of Fick's first law describes a steady flow of matter in a material; Fick's second law describes changes in concentration in time. The diffusion coefficient depends on temperature (Požgaj *et al*. 1997; Hrčka and Babiak 2008) and anatomical direction (Požgaj *et al.* 1997), and it exponentially increases as the moisture content increases, according to studies by Stamm (1959) and Avramidis and Siau (1987). According to Sargent *et al.* (2010), the difference between the heartwood and the sapwood of pine wood is not significant. In addition, the value of the diffusion coefficient depends on the type of boundary condition (Hrčka 2010).

The nonsteady method for determining the diffusion coefficient used in this paper requires the samples to reach their equilibrium moisture content (EMC). The EMC values were used to draft a sorption isotherm for all researched wood species. There are many sorption isotherm models for wood with different approaches and factors (Skaar 1988). The Hailwood–Horrobin model, outlined in a study by Hailwood and Horrobin (1946), is based upon the assumption that a solid solution is formed from a polymer and water. Two or more components can be in equilibrium in this solution. The sorption-isotherm model outlined in a study by Dent (1987) was derived from the theory of surface adsorption, under the assumption that in large internal wood surfaces, the isolated sorption sites are formed by hydroxyl groups. Another approach to the sorption-isotherm model is the ABC isotherm. This type of isotherm substitutes the monolayer moisture content in the equation with the fractional relative humidity (or water activity). It is mathematically equivalent to the Hailwood–Horrobin and Dent models (Zelinka *et al.* 2018).

Some studies were dedicated to investigating on the topic of moisture uptake and sorption isotherms of various thermal insulations and materials. Moisture contents and sorption isotherm of wood fiber thermal insulation were studied by Slimani *et al.* (2019). Sorption of glass wool, rock wool, expanded polystyrene, wood fiber boards and polyester fiberfill was studied in the paper by Ducoulombier and Lafhaj (2017). Water uptake by

various wood species treated with white, brown rot fungi, blue stain fungi, and with various suspensions was presented in the article by Žlahtič and Humar (2017). Water uptake by a material made from fungal mycelium was studied by Haneef *et al.* (2017). Diffusion coefficients of wood-based materials (plywood, OSB, melaminfaced board, particle and fiber board) were studied by Sonderegger and Niemz (2009).

The aim of this paper is to report the diffusion-coefficient measurements, and determine a sorption isotherm for spruce, beech, and oak heartwood decayed *via Trametes versicolor* L. Since wood species is one of the factors affecting the diffusion coefficient (Požgaj, *et al.* 1997); the three before mentioned wood species were chosen for this experiment. *Trametes versicolor* L. is a white rot fungus that primarily degrades lignin, followed by cellulose and hemicelluloses and it was found to decay wood in a simultaneous pattern (Bari *et al.* 2018). The amorphous region of cellulose is much more susceptible to initial fungal metabolism than the crystalline region (Eaton and Hale 1993). Hence, a high rate of cellulose degradation in the initial attack by the fungus would be normal (Bari *et al.* 2018; Bari *et al.* 2019). The cell walls of the wood attacked by this species of fungus are thinned, *i.e.*, all the cell-wall components are degraded. In addition, tiny bore holes are found, which can develop into cell-wall ruptures (Bari *et al.* 2015, 2018). It is obvious that compared to undegraded wood, decayed wood has different structures, chemical compositions, and properties (Bari *et al.* 2018).

Increased porosity could make the sorption process happen faster, since the anatomical structure becomes more porous, there could eventually be even more passageways. Degraded wood is also expected to be able to diffuse more water in comparison to undegraded wood. The equilibrium moisture content at certain relative-humidity ranges is expected to be the same for all three researched wood species. A porous material with a high diffusion coefficient could possibly be used as a building-insulation material for environments with oscillating relative air humidity.

The importance of studying diffusion coefficient and equilibrium moisture content of degraded wood comes from the cited publications. Diffusion coefficient and equilibrium moisture content in degraded wood were not studied yet, and the results of this research will help to understand the sorption kinematics in degraded wood. The results presented in this paper are a part of an experiment on studying thermal and moisture related properties of the selected wood species degraded with *Trametes* versicolor L. The idea of proposing degraded wood as an insulation material originated in the results published by Slováčková *et al.* (2018). Spruce wood degraded with *Trametes versicolor* L. showed lower thermal conductivity than undegraded spruce wood.

EXPERIMENTAL

Materials and Methods

One coniferous and two broadleaved species were chosen for this experiment. Spruce wood (*Picea abies* L.) was chosen, as it is the most widespread coniferous species in Slovakia with a share of 22.45% in the forests. Beech (*Fagus sylvatica* L.) is the first (33.8%) and oak (10.5%) is the second most widespread broadleaved species in Slovakia (Moravčík *et al.* 2018). The lumber used for the beech and sessile oak samples was stored in the Department of Wood Science (Zvolen, Slovakia), and it was obtained from Lesný podnik (a forest space in Zvolen, Slovakia). The spruce lumber was obtained from a window frame manufacturer. For sessile oak samples (*Quercus petraea*, Matt. Liebl.),

the heartwood part of the lumber was used for the samples (in further text, referred to as "oak heartwood").

The average oven-dried densities of the undegraded samples were 394.6 kg·m^{-3} for spruce wood, 688.7 kg·m^{-3} for beech wood, and 730.1 kg·m^{-3} for sessile oak heartwood. The samples did not contain any knots or other defects. When cutting the samples, the grain was always properly aligned with the general anatomical directions. The samples were cut and sanded to a size of 50 mm x 50 mm x 8 mm, with the smallest dimension corresponding to each anatomical direction. A total of 4 samples per anatomical direction per wood species were tested.

The degradation *via Trametes versicolor* was carried out according to STN EN standard 113 (1998). The total degradation time was 6 months, and after this time had passed, the samples were taken out of the Kolle flasks and cleaned of visible mycelium. The samples were then submerged in distilled water, since wood-decaying fungi need a minimum of 5% to 20% air in wood to survive (Rypáček 1957; Reinprecht 2016). Hence, submerging the samples in distilled water stops fungal activity, as it pushes the air out of the wood.

The samples were kept in the water until they had reached maximum moisture content. This was checked regularly by double weighing the wet samples. Each wood species was put into a separate container, and the water was periodically changed. The containers were stored in a dark room at a constant temperature of 20 °C. Once the samples had reached the maximum moisture content, they were left to dry at room temperature for a few days before being oven-dried.

To see the extent of the degradation and structure of the wood after degradation, a few samples were examined under a light microscope. The samples were cut into thin slices on a sledge microtome (Reichert, Wien, Austria) and mounted permanently on a microscope slide with Euparal (BioQuip Products Inc., Rancho Dominguez, United States). Toluidine Blue stain was used to stain beech and sessile oak heartwood samples. Spruce wood was left in its natural state, as the stain made most of the images very dark and the structure of the sample was unclear in the images. The magnification of the microscope was 200x and the camera used for taking images was by Canon, model EOS 600D.

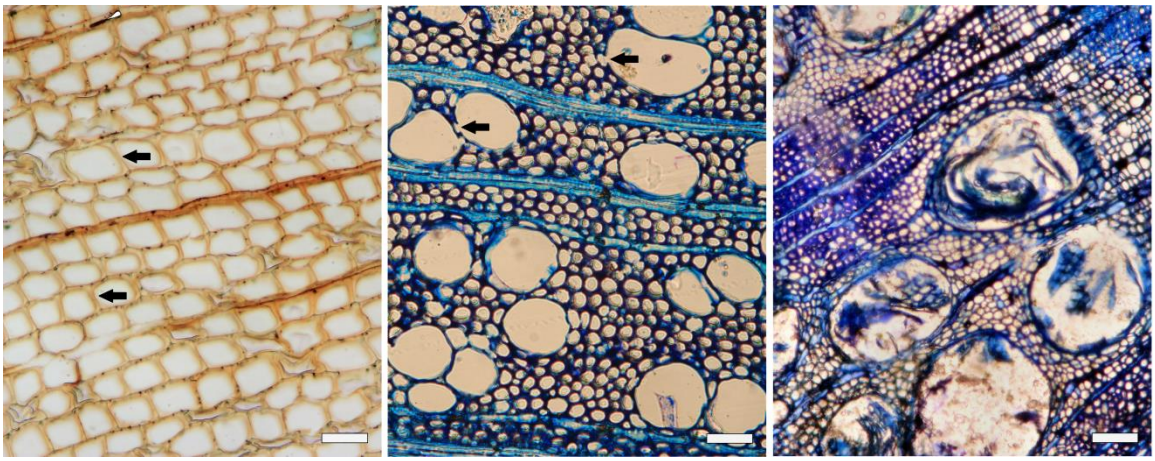

Fig. 1. Images of transversal cuts of degraded spruce (on the left), degraded beech (middle) and degraded sessile oak heartwood (on the right). Scale in the images is 50 µm and it is denoted with a light grey stripe at the lower right corner of each image. The black arrows point at visible cell wall thinning and disruptions.

Images in Fig. 7 show visible cell wall thinning of earlywood tracheids in degraded spruce wood. Vessels in degraded beech wood were not only thinned, but also visibly disrupted in some places. Pits in parenchyma cells of degraded beech wood became very visible through the degradation process. The degradation process is not as apparent in degraded oak heartwood, because the examined sample had a low mass loss. Big tyloses were visible in sessile oak heartwood. Hyphae infestation was visible on some tyloses, but the degradation process of the tyloses is not visible.

The wood moisture content at the cell-wall saturation point was calculated according to the method proposed by Hrčka (2017). This method requires the weighing of the samples in different conditions and environments. First, the submerged samples were weighed *via* a double weighing method after reaching their maximum moisture content (apparent mass in water: m_{zmax}), which is equal to the maximal amount of bound water in the cell walls. The samples were then weighed in an air environment at their maximum moisture content (m_{max}), and then they were oven-dried to calculate the m_0. The maximum free water moisture content (w_{Fmax}) is the moisture content of free water in wood (Hrčka 2017). The maximum moisture content (w_{max}) was calculated according to Eq. 1,

$$w_{max} = \frac{m_{max} - m_0}{m_0} = w_{Fmax} + \frac{m_{zmax}}{m_0} \quad (1)$$

where m_{max} (kg) is the mass of the sample at its maximum moisture content. This mass is determined in the environment of air. m_0 (kg) is the mass of oven-dried sample. w_{Fmax} (-) is the moisture content of the sample at its maximum free water content, m_{zmax} (kg) is the apparent mass of the sample at its maximum moisture content. This mass is determined in the environment of water.

The oven-dried samples were inserted into an air-conditioning (A/C) chamber set to an air temperature of 20 °C ± 2 °C and various relative air humidities (30% ± 3%, 60% ± 3%, and 96% ± 3%). Then the samples were weighed at t = 0, 1, 2, 4, 8, and 24 h from the start of the experiment, and then at t = 7, 21, 28, 35, 42, 49, and 56 d, respectively, until the equilibrium moisture content was reached. Dimensionless concentrations were calculated for all weigh-ins from the moisture content of the samples calculated from their masses according to Eq. 2,

$$c = \frac{w_x - w_0}{w_{EMC} - w_0} \quad (2)$$

where w_x is the moisture content at a certain weigh-in, w_0 is the initial moisture content, and w_{EMC} is the equilibrium moisture content of the sample.

Three relative air humidity stages were set in the A/C chamber: φ = 30% ± 3% (first stage), 60% ± 3% (second stage), and 96% ± 3% (third stage). The samples were not oven-dried between the stages. The dimensions of each sample were measured before and after every stage of the experiment with a Digimatic slide caliper (Absolute, Mitutoyo, Kanagawa, Japan). The A/C chamber was manufactured by Binder (Model KBF 720, Binder GmbH, Tuttlingen, Germany). The laboratory scale was manufactured by Radwag, (Model XA 60/220/X, Radwag, Radom, Poland), with an accuracy of $1 \cdot 10^{-5}$ g.

The control data and the diffusion coefficient results obtained with this method were already previously published in various sources. Diffusion coefficient values for undegraded spruce wood were published in *Štruktúra a vlastnosti dreva* (Structure and Properties of Wood) by Požgaj *et al.* 1997, page 204, Table 10.6. The values for undegraded beech wood were published in the article *Finite Changes of Bound Water Moisture Content in a Given Volume of Beech Wood* by Hrčka in 2015, page 317, Table 1.

Although the previously published experiments were not performed with the same exact relative humidity stages as in this experiment, they were judged to be suitable for usage as control data. The calculation method and proposed weighing steps were the same.

Diffusion-Coefficient Calculation

An inverse method for calculating the diffusion coefficient was used. With regard to this experiment, the dimensionless moisture concentrations of the samples were determined from their masses (as calculated by Eq. 2). The diffusion coefficients were determined for all anatomical directions simultaneously using Fick's second law with a boundary condition of the first kind. This equation was solved as a 3-dimensional problem, as shown in Eqs. 3 and 4,

$$c\,(L,t)\; = f(t) \tag{3}$$

where c_∞ and c_0 are the dimensionless concentrations (c_∞ is the infinite concentration of degraded wood and c_0 is the initial concentration at the start of each stage of the experiment). Both concentrations were calculated according to Eq. 4,

$$c = c_\infty + (c_0 - c_\infty) \cdot \sum_{i=0}^{\infty} \sum_{j=0}^{\infty} \sum_{k=0}^{\infty} \frac{2}{\left((2\cdot i+1)\cdot\frac{\pi}{2}\right)^2} \cdot \frac{2}{\left((2\cdot j+1)\cdot\frac{\pi}{2}\right)^2} \cdot \frac{2}{\left((2\cdot k+1)\cdot\frac{\pi}{2}\right)^2} \cdot$$
$$exp\left[-\left(D_L \cdot (2\cdot i+1)^2 \cdot \left(\frac{\pi}{2\cdot s_L}\right)^2 + D_R \cdot (2\cdot j+1)^2 \cdot \left(\frac{\pi}{2\cdot s_R}\right)^2 + D_T \cdot \right.\right.$$
$$\left.\left.(2\cdot k+1)^2 \cdot \left(\frac{\pi}{2\cdot s_T}\right)^2\right) \cdot t\right] \tag{4}$$

where indices *i*, *j*, and *k* are the dimensionless concentrations of the samples in the corresponding anatomical directions at certain weigh-in times; D_L, D_R, and D_T are the diffusion coefficients in the corresponding anatomical directions; s_L, s_R, and s_T are the dimensions of the samples, with indices L, R, and T denoting the anatomical direction; and *t* is the time when samples were weighed. The two larger dimensions were divided by 2.

The data from Požgaj *et al*. (1997) was used as a base for plotting this equation in MS Visual Basic for Applications. The input data was as follows: the weigh-in time stamps (*t*), the dimensionless concentrations for 1 sample per anatomical direction (*i*, *j*, and *k*), and their dimensions (s_L, s_R, and s_T). The values of the diffusion coefficients (D_L, D_R, and D_T) were estimated. The square-sum deviations were calculated from the experimental and computed data of the dimensionless concentrations, and a sum of squares was calculated. This sum was then approximated to the smallest possible sum with a nonlinear least-squares method in the Solver MS Excel 2010 application. The sum of the squares was set as the objective, and the D_L, D_R, and D_T were set as the changing variable cells.

Sorption-isotherm plotting

The equilibrium moisture content of the samples at all three sorption stages and relative air humidities were used to draft the sorption isotherms for all researched wood species. Dent's sorption-isotherm model was used, as shown in Eq. 5,

$$\frac{w}{w_m} = \frac{b_0\cdot\varphi}{(1-b\cdot\varphi)\cdot(1+b_0\cdot\varphi-b\cdot\varphi)} \tag{5}$$

where *w* is the wood moisture content, w_m is the wood moisture content when the monolayer is completely filled with water molecules, and φ, *b*, and b_0 are the relative air humidity conditions set in the A/C chamber, denoted as temperature functions (Dent 1987).

The square deviations and sum of squares were calculated from the experimental and computed data. The w_m, b, and b_0 values were calculated with the nonlinear least-squares method in the Solver application in MS Excel 2010. The objective cell was the sum of squares, and the changing variable cells were w_m, b, and b_0.

Calculation of the sorption sites

The number of sorption sites was calculated for each species according to Eq. 6,

$$N = \frac{N_A \cdot w_m}{M_{H2O}} \quad (6)$$

where N_A is Avogadro's number, w_m is the moisture content on the monolayer (calculated from the sorption isotherm), and M_{H2O} is the molar weight of water.

Data analysis

The Kruskal–Wallis one-way analysis of variance was used in for statistical data analysis. This is a typical nonparametric method, and it can substitute one-way ANOVA when there is not normal distribution of the residuals and homoskedasticity. The null hypothesis (H_0) was tested in opposition to the alternative hypothesis (H_1) as follows: H_0 - medians are equal in all k subgroups; and H_1 - at least two distributions are different (in terms of medians).

This is a typical signed-rank test where all n values are ranked in a nondecreasing sequence, and this array is analyzed. The sum of the rank in each subgroup $j = 1, 2 \ldots, k$ is marked as T_1. The test statistic of H has *chi*-squared distribution, and is shown in Eq. 7,

$$H = \left[\frac{12}{n(n+1)} \sum_{j=1}^{k} \frac{T_j^2}{n_j}\right] - 3(n+1) \quad (7)$$

where n is the total number of observations across all groups. n_j is the number of observations in group j. T_j is the average rank of all observations in group j.

It is possible to detect concrete pairs with significant differences in the medians with post hoc analysis in case of the rejection of the null hypothesis (H_0). Individual analyses are supported with a graph box-plot presentation of the measured data. The quartile box-plot graphs are shown according to the flow of the used methods.

All analysis was performed in STATISTICA software (Version 12, Tibco Software, Palo Alto, CA). The results of all tests are shown with a significance level of $\alpha = 1\%$. The tables were edited in MS Word 2010, and graphs were left in their original format.

RESULTS AND DISCUSSION

The median values and the first (25%) and third (75%) quartiles of the diffusion coefficients (D_x) for all wood species, anatomical directions, and relative-air-humidity settings are summarized in Table 1. In addition, the control data from previously published experiments are listed. The number of calculations differs for each wood species since the same number of samples available for the measurements was not the same. Some samples were very much damaged due to the high stage of degradation and were deemed unsuitable for calculations.

The D_x values of the degraded spruce wood samples differ from the control D_x values of the undegraded spruce wood. The D_L values at a similar RH were higher, and the

D_R and D_T values of the degraded wood samples were significantly higher than the undegraded wood.

Table 1. Median Values and Quartiles of the Diffusion Coefficients

Wood Species	Relative Humidity (%)	D_L (10^{-10} m^2·s^{-1})	D_R (10^{-10} m^2·s^{-1})	D_T (10^{-10} m^2·s^{-1})	Control Values (10^{-10} m^2·s^{-1})
Spruce (n = 48)	30 ± 3	38.34 (36.84 to 45.61)	6.88 (5.57 to 7.46)	2.04 (1.55 to 2.64)	t = 20 °C; RH = 65% D_L = 40.73 D_R = 2.49 D_T = 1.96 (Požgaj *et al.* 1997)
	60 ± 3	28.94 (24.85 to 32.55)	11.09 (9.29 to 12.52)	7.85 (4.96 to 10.70)	
	96 ± 3	3.37 (2.77 to 3.76)	2.94 (2.58 to 3.14)	2.81 (2.57 to 3.12)	
Beech (n = 80)	30 ± 3	47.93 (39.09 to 56.56)	11.76 (8.90 to 13.11)	3.18 (1.92 to 4.70)	t = 20 °C; RH = 85% D_L = 15.10 D_R = 0.45 D_T = 0.20 (Hrčka 2015)
	60 ± 3	31.22 (29.65 to 33.64)	8.98 (6.73 to 9.67)	2.94 (2.33 to 4.35)	
	96 ± 3	3.46 (3.17 to 3.83)	1.70 (1.46 to 1.94)	1.56 (1.32 to 1.71)	
Sessile Oak heartwood (n = 64)	30 ± 3	6.60 (4.63 to 9.62)	0.39 (0.16 to 1.81)	4.02 (3.02 to 4.94)	(no control data)
	60 ± 3	4.99 (4.00 to 10.70)	0.47 (0.19 to 1.40)	3.08 (2.42 to 3.88)	
	96 ± 3	1.49 (1.02 to 2.13)	0.41 (0.31 to 0.51)	1.2 (1.10 to 1.31)	
Note: Quartiles are in Brackets (Q1 through Q3); For RH, the air temp = 20 ± 2 °C					

A comparison of the D_x values of the control and degraded samples in the transversal directions for beech wood showed a similar result to the spruce wood samples. The control D_L value was similar, although slightly higher, than the undegraded wood. The D_x values of the control samples in the transversal direction were significantly lower at a RH of 85% than the D_x values of degraded wood at a higher RH (96%). Following the median values trend for the D_x values of the degraded wood samples, it is possible to conclude that degraded beech wood displays higher D_x values in the transversal direction.

The diffusion coefficients of sessile oak heartwood displayed great variability. Some calculations of the sessile oak heartwood yielded a zero value for the D_R. This may have been due to variability in weight loss in these samples; in some, there was weight loss of approximately 10%, whereas the rest of the samples showed an average weight loss of approximately 40%. The median mass losses were as follows: 45.81% for degraded spruce wood, 45.00% for degraded beech wood, and 31.40% for sessile oak heartwood. The variation coefficients of mass losses were 12.29% for degraded spruce wood; 15.98% for

degraded beech wood and 48.70 % for degraded sessile oak heartwood. A further statistical evaluation on the weight loss of the samples was not performed, as there were not enough samples for a thorough evaluation.

Since there were no control data for sessile oak heartwood, the authors compared the data to the work of other researchers. Chen *et al.* (1994) researched the D_x at an air temperature of 43 °C and a RH of 45% with an initial sample EMC of 12%. They proposed D_x values of $14.08 \cdot 10^{-10}$ $m^2 \cdot s^{-1}$ in the longitudinal direction and $3.85 \cdot 10^{-10}$ $m^2 \cdot s^{-1}$ in the transversal direction for red oak heartwood. The experiment by Wang and Cho (1994) was performed on 0.5 cm thick samples with the sorption conditions as follows: an air temperature of 20 °C and a RH of 90%. The proposed D_x values for the red oak wood samples were $6.2 \cdot 10^{-11}$ $m^2 \cdot s^{-1}$ in the longitudinal direction, $1.8 \cdot 10^{-11}$ $m^2 \cdot s^{-1}$ in the radial direction, and $0.7 \cdot 10^{-11}$ $m^2 \cdot s^{-1}$ in the tangential direction. Peralta and Bangi (2003) researched variation in the D_x values during a drying process with three stages. The value for the longitudinal direction was approximately $2.34 \cdot 10^{-9}$ $m^2 \cdot s^{-1}$, and the values in the radial direction were $9.16 \cdot 10^{-11}$ $m^2 \cdot s^{-1}$, and $7.75 \cdot 10^{-10}$ $m^2 \cdot s^{-1}$, with respects to the 3 stages of stepwise drying, from green to an EMC of 8%. The D_x values of the degraded oak heartwood samples presented in this paper were closest to the findings of Chen *et al.* (1994). The D_x values of the degraded oak heartwood in the tangential direction were similar, but the values in the longitudinal direction were lower.

According to Levene's test, the 3 factors of the diffusion coefficients, *i.e.*, wood species, anatomical direction, and relative air humidity, showed that the variances were not equal (as shown in Table 2).

Table 2. Levene's Test Results

P Variances	Levene F (1 df)	df Levene	p Levene
369.4516	2.697016	136.9853	0.00
Note: F_{crit} = 26.1647			

The Kruskal–Wallis one-way ANOVA showed no significant difference between the D_x values of the degraded spruce and beech wood samples. However, the results were statistically significant for the D_x comparisons between the spruce and sessile oak heartwood samples, and the beech and sessile oak heartwood samples. These results are presented in Fig. 2.

Further statistical comparison of the D_x *via* the Kruskal–Wallis one-way ANOVA was performed, with the anatomical direction as the factor, and showed difference between the longitudinal and both transverse directions. However, there was no statistically significant difference between the radial and tangential directions (Fig. 3). It must be noted, that all researched degraded wood species were evaluated in this comparison with the factor anatomical direction.

Similar results were obtained from the comparison of the D_x with relative air humidity as the factor. All researched wood species were included in this comparison. There was no significant statistical difference between the D_x values calculated with relative humidity levels of 30% ± 3% and 60% ± 3%. However, a significant statistical difference was found between the D_x values calculated with a RH of 30% ± 3% and 96% ± 3%, and the D_x values calculated with a RH of 60% ± 3% and 96% ± 3%. These results are displayed in Fig. 4.

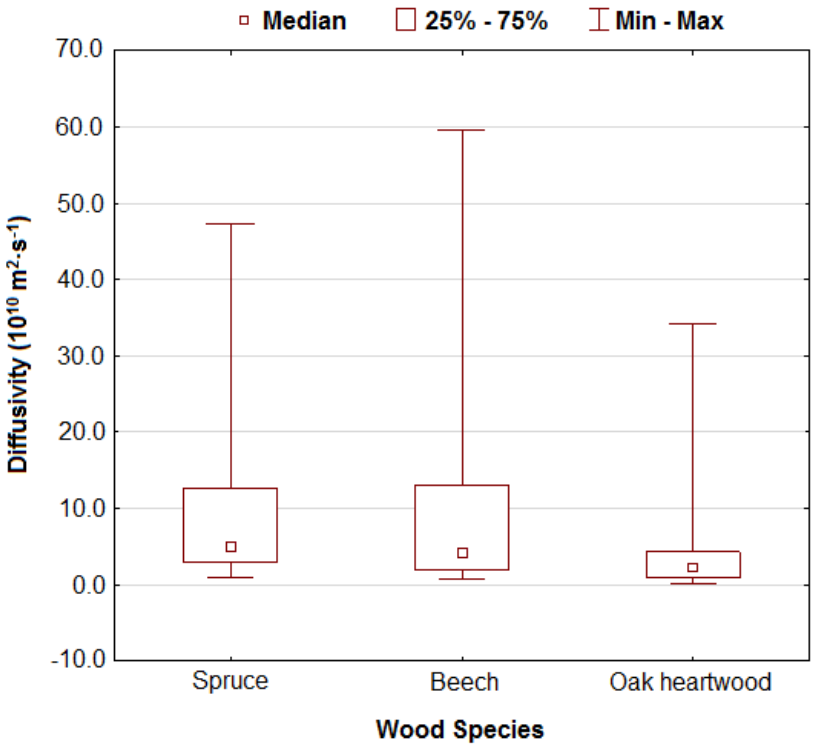

Fig. 2. Box-plot graph of the diffusion coefficients of the researched degraded wood species; factor: wood species

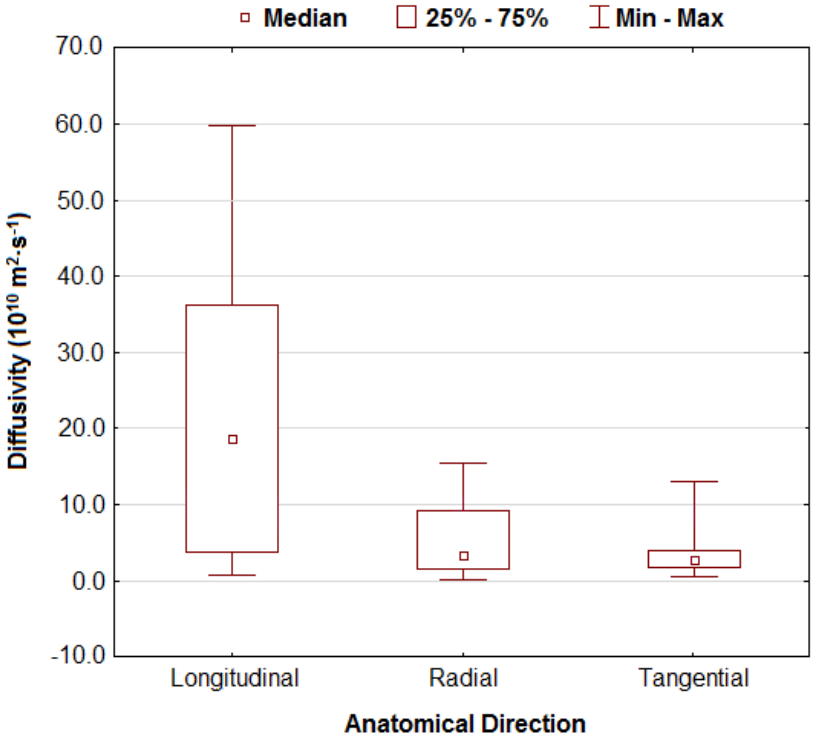

Fig. 3. Box-plot graph of diffusion coefficients of the researched degraded wood species; factor anatomical direction

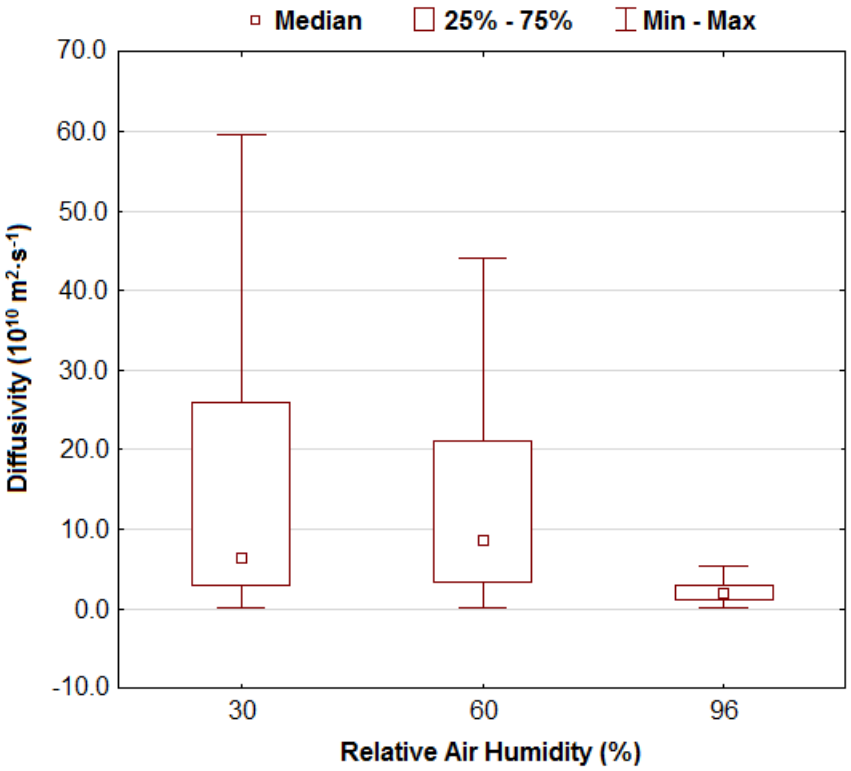

Fig. 4. Box-plot graph of diffusion coefficients of the researched degraded wood species; factor relative humidity

According to studies by Stamm (1959) and Avramidis and Siau (1987), the D_x value increases as the RH increases. However, this relationship is not the same for degraded wood, according to findings in this paper. The median values of the D_x decreased as the RH increased, and sharply dropped with a high RH.

These results show that degraded wood (and wood in general) quickly reacts to changes in relative air humidity. Wood can absorb and desorb a large amount of water in a short amount of time. From the researched wood species, the degraded beech wood samples were found to have larger D_x values than the undegraded beech wood samples, which meant it could absorb water faster.

With regards to the sorption-isotherm results, the Levene's test performed on the equilibrium moisture content values of all samples did not show variance in the samples. However, Duncan's multiple-range test showed differences when the wood species was used as the factor, so a sorption isotherm had to be individually plotted for each wood species (as shown in Figs. 5 to 7).

The w_m, b, and b_0 values, as well as the calculated sorption sites (N) are presented in Table 3. The spruce wood samples displayed slightly higher moisture content values in the monolayer than the beech and oak heartwood samples did. According to Požgaj *et al.* (1997), the average moisture content in the monolayer ranged from 5% to 8%. Functions b and b_0 had similar values for all wood species samples. The degraded spruce wood samples were found to have the most sorption sites among the researched wood species.

Table 3. Calculated Values of the w_m, b, and b_0 Characteristics of Dent's Isotherm

Tree Species	w_m (%)	b	b_0	N (kg^{-1})
Spruce	9.46	0.73	3.53	$3.16 \cdot 10^{24}$
Beech	8.61	0.75	3.35	$2.88 \cdot 10^{24}$
Sessile Oak heartwood	8.55	0.73	3.34	$2.86 \cdot 10^{24}$

The sorption isotherms for each researched wood species are presented in Figs. 4 to 6. The experimental values were in good agreement with the calculated values for all 3 stages of the sorption experiment (Table 4). The values for the cell-wall saturation limit at a relative air humidity of 100% were calculated from the sample masses in different environments, accordingly to Eq. 1.

The cell-wall-saturation (CWS) limit and EMC at RH of 100% calculated from the isotherm was in agreement only with the spruce wood samples; the beech and sessile oak heartwood samples showed discrepancy. The reason for this could be the leaching of a portion of the contents during the sterilization of the samples in distilled water. The distilled water that the samples were submerged in was always colored before being changed.

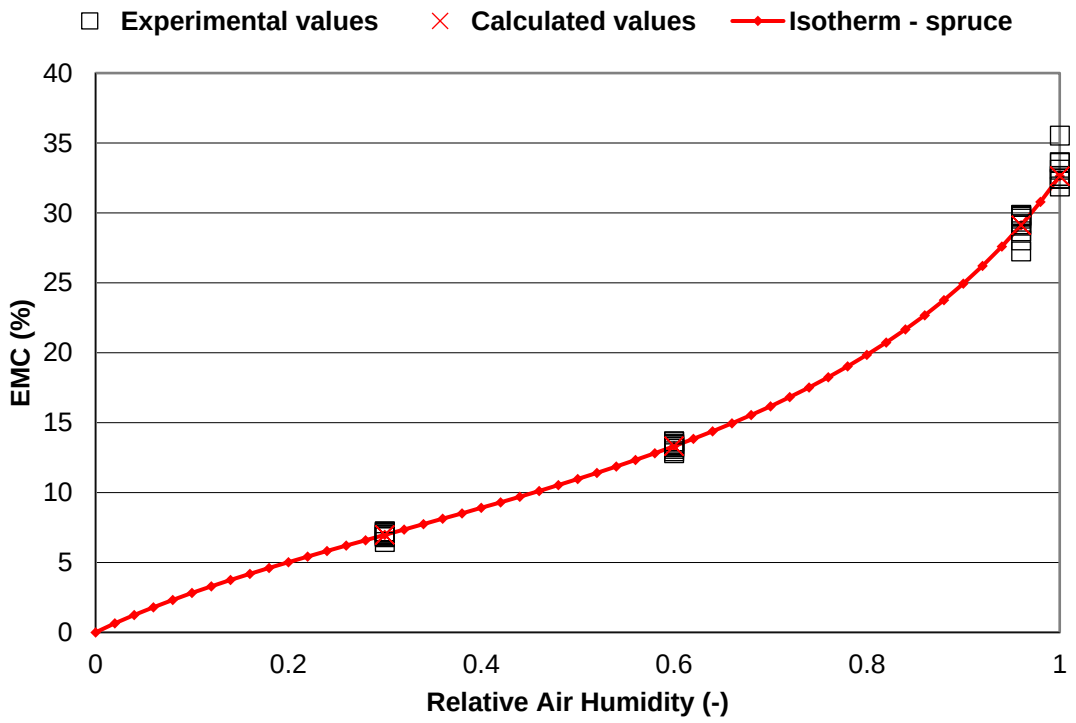

Fig. 5. Sorption isotherm of degraded spruce wood

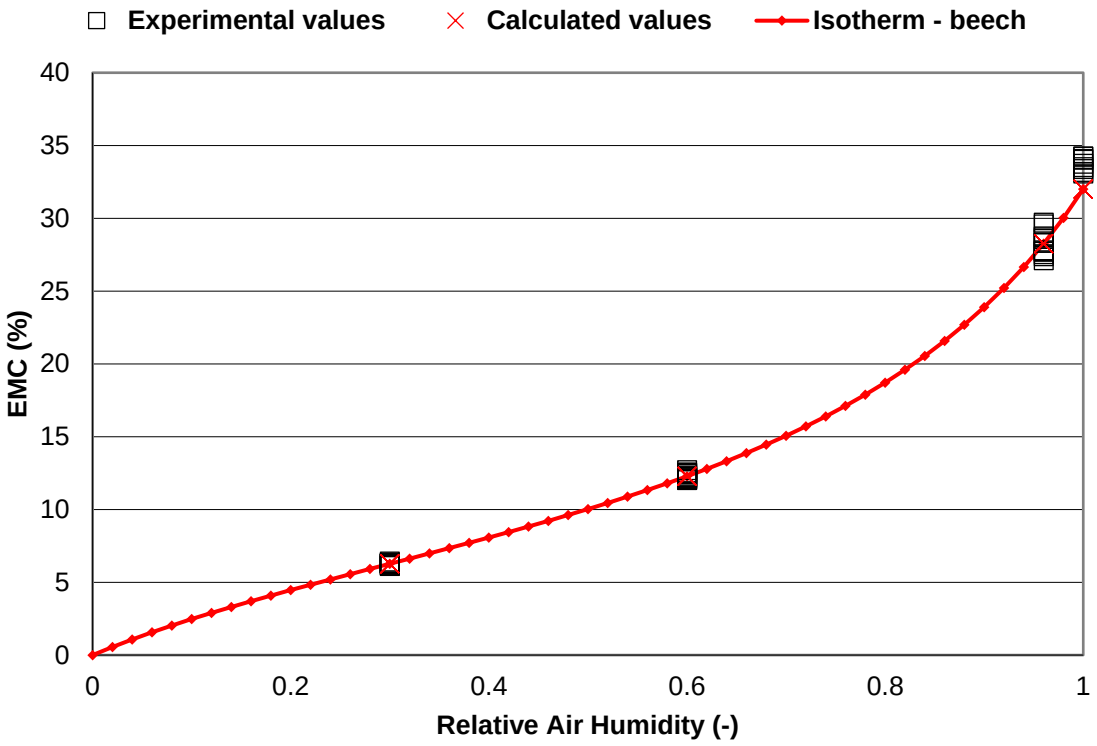

Fig. 6. Sorption isotherm of degraded beech wood

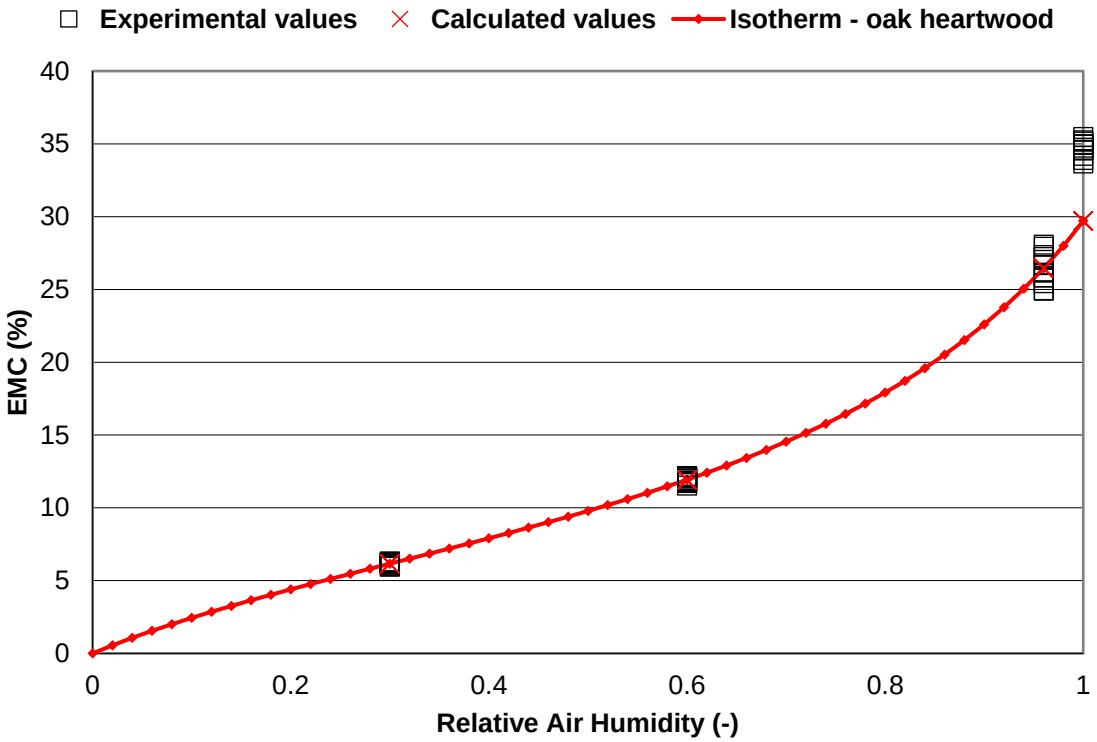

Fig. 7. Sorption isotherm of degraded sessile oak heartwood

Comparison data to a similar research on sorption isotherm of a wood-based material can be found in the research by Slimani *et al.* (2019). The researched material was wood fiber insulation. The moisture contents for relative air humidity stages were (in the process of sorption): 4.88 % for RH = 30%; 8.08% for RH = 60% and 20.26% for RH = 97% (Slimani *et al.* 2019). The moisture contents of wood fiber insulation for the certain relative air humidity stages were lower (by a few percent) than moisture contents reached by degraded wood species at each relative humidity stage. It must be noted, that wood fiber insulation is usually treated with fire retardants which could have an impact on the sorption properties.

Another study on water uptake by a material created from fungal mycelium was presented by Haneef *et al.* (2017). Mycelium of *Pleurotus ostreatus* and *Ganoderma lucidum* were fed two types of substrates – cellulose and cellulose/potato dextrose. The graph of the water uptake by the prepared materials has a similar shape to the sorption isotherms of wood. The water uptake at RH = 100% was around 12% for three out of four materials. The water uptake for the fourth material was below 20% at RH = 100% (Haneef *et al.* 2017). The water uptakes of the materials presented in the paper of Haneef *et al.* (2017) were all lower than the moisture contents of the degraded wood species researched in this paper.

There is also a study of sorption for glass wool, rock wool, expanded polystyrene, wood fiber board and polyester fiberfill by Ducoulombier and Lafhaj (2017). This experiment was done at a slightly higher temperature of 23 °C in the A/C chamber and the relative air humidity stages were set to 50, 80, 93, and 97%. The materials were also tested by immersion in water. It was determined that rock wool and polyester fiberfill did not absorb water even at RH = 97% (Ducoulombier and Lafhaj 2017). Glass wool absorbed a small amount of water. Wood fiber board had a moisture content of around 26% at RH = 97% (Ducoulombier and Lafhaj 2017), which is close to the moisture content reached by the degraded wood species researched in this paper. Wood fiber boards reached a moisture content of around 260% after immersion in water (Ducoulombier and Lafhaj 2017), which is close to the maximum moisture content reached by degraded beech wood in the present experiment (Table 4).

Table 4. Oven-dried Densities, Cell-wall Saturation Moisture Contents and Maximum Moisture Contents of Degraded Wood Species

Condition	Degraded Spruce Wood	Degraded Beech Wood	Degraded sessile Oak Heartwood
Oven-dried density ($kg \cdot m^{-3}$)	200.3	341.8	512.9
Cell-wall saturation (CWS) (%)	32.64	33.79	34.85
Maximum moisture content (%)	464.98	256.74	164.09

Table 4 shows the oven-dried density median, the CWS, and the maximum moisture content of all researched wood species. The CWS was similar for the spruce and beech wood samples, and the sessile oak heartwood samples had the highest CWS. Degraded spruce wood samples had the lowest density and the highest maximum moisture content, while the sessile oak heartwood samples had the highest density and the lowest maximum moisture content.

Water uptake in wood occurs in the hygroscopic (RH ranges from 0% to 95%) or overhygroscopic (RH ranges from 95% to 100%) range (Espinosa and Franke 2006; Nilsson *et al*. 2018). Water uptake in the hygroscopic range has an EMC ranging from 0% to 30%. Wood can absorb greater than 150% water when the RH ranges from 99.5% to 100% (Stamm, 1964; Fredriksson 2019). As such, the degraded spruce and beech wood samples can absorb far more water within a RH ranging from 99.5% to 100%.

CONCLUSIONS

1. Analysis of the factors affecting the diffusion coefficients showed that the wood species, relative humidity, and anatomical direction had some differences. The difference was significant between the values of the spruce and sessile oak heartwood samples, and between the beech and sessile oak heartwood samples.
2. Analysis of the relative humidity factor showed that there was no significant difference between the diffusion coefficients of samples under relative-humidity ranges of 30 ± 3% and 60 ± 3%. The decrease in the diffusion-coefficient values was significant in the 96 ± 3% relative humidity range. The diffusion coefficients of the degraded wood samples did not increase with an increasing relative humidity.
3. The diffusion-coefficient value in the longitudinal direction was significantly different from the values in the transversal directions.
4. The results show that degraded wood quickly equilibrates to moisture-related changes in the environment in which it is kept. The diffusion coefficients of the degraded wood samples appear to be higher than the diffusion coefficients of the undegraded wood samples. It is apparent that degraded wood can diffuse more water than undegraded wood can.
5. Research on the sorption isotherms showed that the wood species was a significant factor; hence, the sorption isotherm was plotted separately for each wood species. The experimental equilibrium-moisture content values were mostly in good agreement with the calculated values.
6. The results of the diffusion coefficient, equilibrium moisture content, and sorption-isotherm parameter calculations are helpful in determining and predicting the behavior of this material in various relative-air-humidity conditions. Moreover, wood is a natural and ecological material, and wood products should be used more often as building materials.

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