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Research on Determination of Water Diffusion Coefficient in Single Particles of Wood Biomass Dried Using Convective Drying Method

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Abstract: Determination of the mass diffusion coefficient for dried, inhomogeneous material is difficult as it depends on the drying agent temperature and the moisture content and physical structure of the material. The paper presents a method for efficient determination of the water diffusion coefficient for wood solid cuboids dried using convective drying methods. In this work, the authors define a theoretical dependence of the Fourier number on reduced water content in a convectively dried cuboidal solid, based on a simplified theoretical solution of the diffusion equation for such a body. The material for drying included shoots of common osier, robinia (false acacia), multiflora rose, and energy poplar, dried at temperatures of 40, 50, 60, 70, and 80 °C, in free convection. The obtained results differ from the theoretically anticipated changes of the coefficient.

Keywords: diffusion coefficient; drying; convection; wood biomass



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1. Introduction

1.1. General Characteristics of Wood Biomass

Nowadays, much attention is paid to the identification of different species of biomass that can replace conventional energy sources such as hard coal, lignite, or crude oil. There are a few reasons why there has been a growing interest in biomass as an energy source [1–5]. This is mainly due to technological changes related to conversion and to the use of biomass as fuel for lower costs and higher conversion capacity [6,7]. Lignocellulosic biomass is a highly available renewable fuel that is considered to not contribute to the greenhouse effect. In this context, it is expected that biomass and biowaste conversion will play a major role in the global energy transition [8,9]. Wood biomass, a by-product of wood production and various technologies of wooden element production, may be a valuable biofuel. In-depth knowledge of mechanisms and processes that occur during combustion is of primary importance in order to develop more efficient combustion systems [10]. The most important parameters of biomass for energy purposes are the heat of combustion and heat value [11]. These parameters are predominantly related to the chemical composition of the material as well as moisture content.

Wood is composed of various chemical compounds, including cellulose, lignin, hemicelluloses, resins, and gums, as well as water-soluble and water-insoluble mineral compounds. Cellulose is the main component of cell walls. It is a polysaccharide with the general formula of $(C_6H_{10}O_5)_n$ and a crystalline structure. It consists of glucose molecules. It has an influence on the strength and elasticity of wood. Cellulose content ranges between 45% and 55%, depending on the species. Lignin is a binder that builds a skeleton of a cell

wall by mechanically and chemically binding with cellulose [12]. It makes wood hard and stiff. Lignin content in wood ranges between 25% and 30%. Hemicelluloses are polysaccharides that make up the skeleton of a cell wall. Hemicellulose is a polysaccharide composed of D-glucose, D-galactose, D-mannose, D-xylose, and L-arabinose, as well hexuronic acids, namely glucuronic, galacturonic, and methyl glucuronic acids [13]. They can be found in the lignified plant tissues. They serve as spare substances, and their chemical composition and structure are similar to those of cellulose. The content of hemicelluloses in wood ranges between 10% and 20%, depending on the species of the tree. Resins and gums are water-insoluble, shapeless or partially crystalline mixtures of nonvolatile organic and phenolic compounds. They constitute 4.5–9.5% of wood structure, while the content of mineral compounds is between 0.2% and 2.3% [14–16].

Wood biomass for energy purposes usually has to wait a certain period of time to be processed. During that time, its chemical composition may be adversely affected due to the development of microorganisms and fungi, which eventually leads to deterioration and mold formation. Therefore, it is essential that water content in the biomass during its storage be low. This can be achieved by drying to make it suitable for processing. The purpose of drying is to lower the water content to a level that will inhibit enzymatic processes and prevent the growth of microorganisms during storage [17,18].

Wood drying is a complex process that involves penetration and conduction of heat as well as internal transport and rising of steam. There are numerous methods for drying biological materials. However, so far, none have been examined thoroughly enough to be able to effectively predict the dynamics of the process and control it. Controlling the process of drying is extremely important in regards to its optimization. This has been confirmed by research work [19] focused on the optimization of supplying heat energy to the material being dried. Yuan et al. [20], in the research and simulation performed, presented the dynamics of the process of drying a porous body. While the process of drying a single element is less complex, drying a layer of material is more complicated, as heating and cooling of certain sublayers of the material occur simultaneously [21]. Fragmented (size-reduced) wood biomass is stored and dried in a thick layer, which may result in energy and mass losses related to the biomass [22]. Material may be dried using the natural convection method, which is used for technological purposes when necessary (it generates additional cost) [23].

Obtaining knowledge about drying solids, including wood, involves answering two basic questions, namely how a single particle dries (consistent object of wood) and how a bed of such particles dries. Answering the second question is of particular importance for practical reasons. However, this question is difficult to answer as there is no simple answer to the first one.

Due to the morphological structure of wood, the process of water transport in single particles being dried is specific and differs from other materials with a capillary-porous structure. Not only does temperature have an impact on the drying rate, but so does the relative moisture content of the material being examined [24]. Both methods and parameters of drying significantly affect the quality of the dried material [25].

Gonzalez Martinez et al. [26] pointed at the characteristic time analysis of physical and chemical processes involved in torrefaction. They found that heat transfer phenomena are faster than the chemical reaction, which is the limiting factor of the transformation. Generally, the chemical regime is achieved at particle-scale, but this is not always the case at bed-scale. Limitations may appear when bed thickness becomes too high. Moreover, Dupont et al. [27] reported that pyrolysis of biomass seems to occur in a chemical regime for small particles (<100 mm). In other cases, the transformation seems to be controlled both by the heat transfer and the chemical kinetics.

Wood has a capillary-porous structure. The main thermodynamic forces that are responsible for water movement in wood are capillary forces and diffusion. Physically, wood drying involves a set of processes that result in liquid water being changed into steam, which speeds up its transport outside.

Another physical phenomenon, sorption, also based on the diffusion of water contained in the air inside dry material, plays a reverse role (Figure 1) [28].

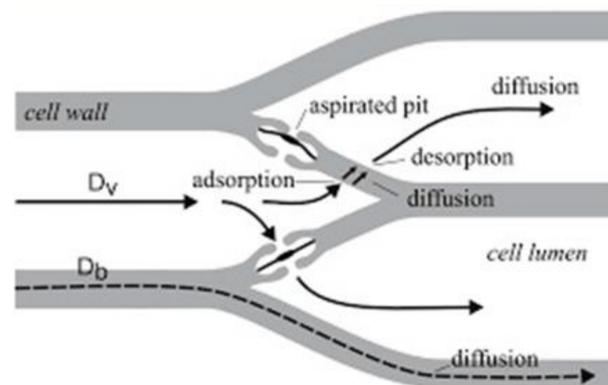


Figure 1. Water diffusion in wood [29].

In the process of drying porous-capillary solids, water is mostly removed via capillaries (Figure 1). Capillaries in wood have diameters of various dimensions and are interconnected. Water flows from capillaries with greater diameters to those with smaller ones, using capillary pressure differences caused by the variation in the diameters. Some energy is used to overcome forces of friction. While drying wood, capillaries filled with water are located next to spaces filled with air, which is also responsible for the movement of water in wood. Capillary forces are responsible for the transport of water from intercellular spaces to the surface of the material. The rest of the trapped water moves via diffusion phenomenon.

Water is the main component of all plants, including trees. Three forms of water may be distinguished in wood from a recently cut down tree [30]:

- Free water—contained in the capillaries, does not have an impact on the dimensions (size) of the material. Evaporation heat is used to evaporate this form of water.
- Bound or hygroscopic water—contained in pores between micelles and bound via hydrogen bonds. In order to evaporate this form of water, the heat of vaporization is used to break the hydrogen bonds.
- Constitutional or capillary water—contained in cell walls, forms organic compounds, and cannot be removed by physical means. Therefore, it is not taken into account when measuring the moisture content of the material and during drying [30].

Wood of high moisture content is not suitable for processing or for use in industry. Hence, its value is low. The process of wood drying results in the refinement of this material [31]. For example, in the case of pellet production, the most common humidity range is 10–15% [32–35].

1.2. Water Diffusion in the Process of Wood Drying

Diffusion is a spontaneous and chaotic movement of molecules and atoms of various substances as a result of intermolecular pressures. Generally, diffusion occurs in liquid and gaseous substances and involves molecular leveling-off of the concentrations of mixture components. As the pressure difference increases on the border of phases of different substances, the process speeds up. Diffusion is also responsible for changes in the liquid content of capillary-porous solids, particularly for wet wood. As in the case of gases and liquids, molecules move from places where concentration is high to places where it is low. High temperatures and low pressure that have an impact on the molecules intensify the process.

In the literature on drying, first-type boundary conditions are almost always taken into account due to the fact that it is easier to obtain the solution using such boundary

conditions. For third-type boundary conditions, the Biot number is necessary; the infinite series constitute the appropriate solutions [36].

In biological materials, especially with high initial moisture content, water can be transported by water diffusion, vapor diffusion, Knudsen diffusion, internal evaporation and condensation effects, capillary flow, and hydrodynamic flow. Often, there is a mixture of various transport mechanisms, and the contributions of the different mechanisms to the total transport vary from place to place and change as drying progresses [37]. Therefore, the water diffusion coefficient in the model of the drying describes the total transport of moisture and is called the effective diffusivity.

There is no diffusion theory that is sufficiently well formulated and verified. Therefore, the most commonly used method for determining the moisture diffusion coefficient is by fitting the diffusion-based drying equations to the experimental data in the drying.

The process of diffusion of the component in the liquid flux is described for the isothermal and isobaric conditions by the so-called empirical Fick's first law, expressed by the following formula:

$$J = -D \frac{\partial \phi}{\partial x} \quad (1)$$

where J is the flux of component (amount of substance which passes per given unit of time through a unit section) ($\text{kg}/\text{s m}^2$), D is the diffusion coefficient (m^2/s), ϕ is the concentration (amount of substance per given unit volume) (kg/m^3), and x is the coordinate of the axis of diffusion direction.

The mass diffusion coefficient is defined as the amount of diffusing component per unit area perpendicular to the considered direction of diffusion, per unit time. It is a quantity defined for a diffusing substance, dependent on the size and shape of the molecule, as well as the pressure and temperature [38]. The mass diffusion coefficient is a parameter in the simplified form of Fick's second law:

$$\frac{\delta \varphi}{\delta t} = D^2 \varphi \quad (2)$$

where D is the mass diffusion coefficient (m^2/s), φ is the substance concentration (mol/m^3), r is the distance gradient from the source of diffusing substance (m), and t is time (s).

Hardwood has thicker cell walls and greater density in comparison to softwood. It is more susceptible to faults such as change of color or cracks during drying. The greater the density of wood, the longer it takes for the wood to dry. This is caused by the fact that it is more difficult for the water or steam moving via diffusion to escape from wood with higher density. During drying, lumens of wood cells approximately retain their size, while the spaces between micelles and cell walls become smaller. This results in the wood shrinking and the water or steam that moves facing greater resistance [30].

Equation (2) indicates that more thorough knowledge of the process of drying capillary-porous materials, including wood, can be obtained by defining the value and learning the changeability of the coefficient of the internal mass diffusion. The geometric shape (infinite plate, cylinder, ball, etc.) of the dried material has an influence on the determination of the water diffusion coefficient [39]. In practice, however, the method to determine its value is a problem [40], as there are no direct methods to measure the water diffusion coefficient in non-homogeneous, moist solids. Indirect methods may be applied to determine the coefficient. These methods are based on the idea of the inverse problem method, which involves the transformation of theoretical models containing the coefficient. Usually, these models only approximately describe transport of water, a result of the diffusion phenomenon. Hence, the calculated coefficient may be named the "conventional" or "effective" water diffusion coefficient. The effective water diffusion coefficient may also be determined very accurately with the inverse problem method, using the finite element method [41].

2. Objective of the Work

The goal of this work was to determine an effective water diffusion coefficient in the process of convective drying. The selection of particles was determined by the popularity of types of wood biomass energy used in Poland, namely osier, robinia, multiflora rose, and poplar, depending on the temperature of drying air.

Due to the non-homogeneous structure of biomass particles being dried, an indirect method was applied to determine the mass diffusion coefficient, based on theoretical models of kinetics of drying. In this work, approximation by a cuboid was used to approximate the volume of biomass particles in the shape of cylinders, which made it possible to use a mathematical model of drying solid cuboids to determine the water diffusion coefficient.

3. Methodology

3.1. Drying Kinetics Measurements

Material for research consisted of shoots of energy plants, i.e., common osier (*Salix viminalis* L.), robinia (*Robinia pseudoacacia*), multiflora rose (*Rosa multiflora* Thunb.), and poplar (*Populus* L.). The samples were collected from the experimental field of the Warsaw University of Life Sciences (SGGW) in Warsaw. Common osier, robinia, and multiflora rose were collected in September, and poplar was collected in November. The sprouts with a diameter of 0.03 m were used to prepare samples, each of which consisted of five 0.05-m-high cylinders. Samples were dried in free convection conditions, at five temperatures, namely 40, 50, 60, 70, and 80 °C, and each drying experiment was repeated thrice [42–45].

Measurements were conducted at a measurement stand in the drying laboratory of the Department of Fundamental Engineering of Warsaw University of Life Sciences, equipped with a convective drier, a computer system recording changes in mass of the samples being dried, and a system for temperature measurement.

3.2. Determination of the Mass Diffusion Coefficient

As there is no method to directly determine the water diffusion coefficient in wood, an indirect method was applied. With this method, the internal water diffusion coefficient may be determined using the theoretical Fourier number from Equation (3) [46]:

$$Fo_m = \frac{a_m \tau}{R^2} \quad (3)$$

where Fo_m is the Fourier number, a_m is the water diffusion coefficient (m^2/s), τ is time (s), and R^2 is the characteristic dimension (m).

The Fourier number is an argument for a simplified solution of the mass diffusion equation, written in the form of Fick's law (Equation (2)), which may also be given as follows:

$$U(\tau) = \frac{u - u_r}{u_0 - u_r} = f(Fo_m) \quad (4)$$

where U is the reduced water content (–), u is the water content in moist body ($\text{kg H}_2\text{O}/\text{kg dry substance}$), u_r is the equilibrium water content ($\text{kg H}_2\text{O}/\text{kg dry substance}$), and u_0 is the initial water content ($\text{kg H}_2\text{O}/\text{kg dry substance}$),

If measurements allow the determination of changes in water content in the body being dried, and the values of $U(\tau)$ in particular, and the structure of relation $f(Fo_m)$ is known, then the determination of the Fourier number, and, subsequently, the water diffusion coefficient in the material being dried, is a question of using an appropriate algorithm of inverse calculations:

$$a_m = \frac{Fo_m \cdot R^2}{\tau} = \frac{f^{-1}(U) \cdot R^2}{\tau} \quad (5)$$

In this work, a model of convective drying of a solid was used as an analytical solution of Equation (2) at the initial ($u(\tau = 0) = u_0$) and boundary ($u|_A = u_e$) conditions of the first type, with reference to mean moisture content as a function of time for a cuboidal body, in the following form [47,48]:

$$U(\tau) = \frac{\bar{u}(\tau) - u_e}{u_0 - u_e} = \frac{512}{\pi^2} \sum_{l=0}^{\infty} \sum_{m=0}^{\infty} \sum_{n=0}^{\infty} \beta_{l,m,n} \exp\left[-\frac{\pi^2}{4} \left[(2l+1)^2 K_1^2 + (2m+1)^2 K_2^2 + (2n+1)^2 K_3^2\right] Fo_m\right] \quad (6)$$

where K_i is the drying rate coefficient, $i = 1, 2, 3$, (1/h); l , m , and n are summing indices relative directions of characteristic dimensions of a cuboid; and $\beta_{l,m,n}$ is the mass conductivity coefficient in the boundary layer of bipermeable surface (m/s). The β coefficient is obtained from the following formula:

$$\beta_{l,m,n} = \frac{1}{(2l+1)^2 (2m+1)^2 (2n+1)^2} \quad (7)$$

The drying rate coefficient may be obtained from the following formula:

$$K_1 = \frac{R}{w}, \quad K_2 = \frac{R}{t}, \quad K_3 = \frac{R}{h} \quad (8)$$

where w , t , and h are the characteristic dimensions of a cuboid (half of length, height, and width) (m) and R is the generalized characteristic dimension (m), expressed by the following formula:

$$\frac{1}{R^2} = \frac{1}{w^2} + \frac{1}{t^2} + \frac{1}{h^2} \quad (9)$$

The dimensions and the shape of cylindrical samples with a length of 0.05 m and a diameter of approx. 0.03 m were transformed to the cuboidal form with the dimensions 0.027 m \times 0.027 m \times 0.05 m (length). The dimensions of the cuboid were selected so that the volume was similar to the volume of a cylinder. The determined characteristic dimensions and shape coefficients are presented in Table 1.

Table 1. Characteristic dimensions and shape coefficients.

w (m)	t (m)	h (m)	K_1	K_2	K_3	R
0.0135 \pm 0.002	0.0135	0.025	0.6606	0.6606	0.3567	0.891

4. Results

Results of measurements of weight changes during drying were converted into water content relative to drying time (Figures 2 and 3).

Equation (6), with a_m determined using the proposed method, describes the drying process well, because the estimated RMSE error determined is very small.

From Equation (5) it is possible to calculate theoretical water contents U , by using the values of the Fourier number from the interval $\langle 0; +\infty \rangle$. As value U , calculated from a series (5), rapidly converges to 0, the calculations were limited to a limited interval, $Fo_m \in \langle 0; 4.9 \rangle$. The graph of the Fourier number (Fo_m) versus reduced water content (U), for a cuboidal body, for several dozen values of Fo_m , is presented in Figure 4.

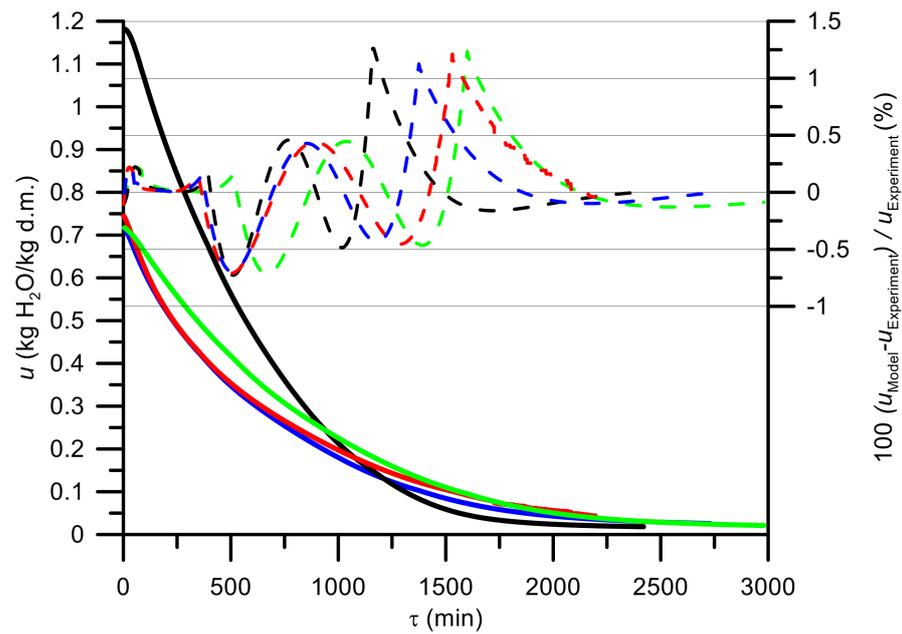


Figure 2. A graph of moisture content versus time of osier (—), rose (—), poplar (—), and acacia (---) drying at 60 °C and results of mathematical modeling of osier (--- RMSE = 0.0007), rose (--- RMSE = 0.0006), poplar (--- RMSE = 0.0011), and acacia (--- RMSE = 0.0007) drying at 60 °C.

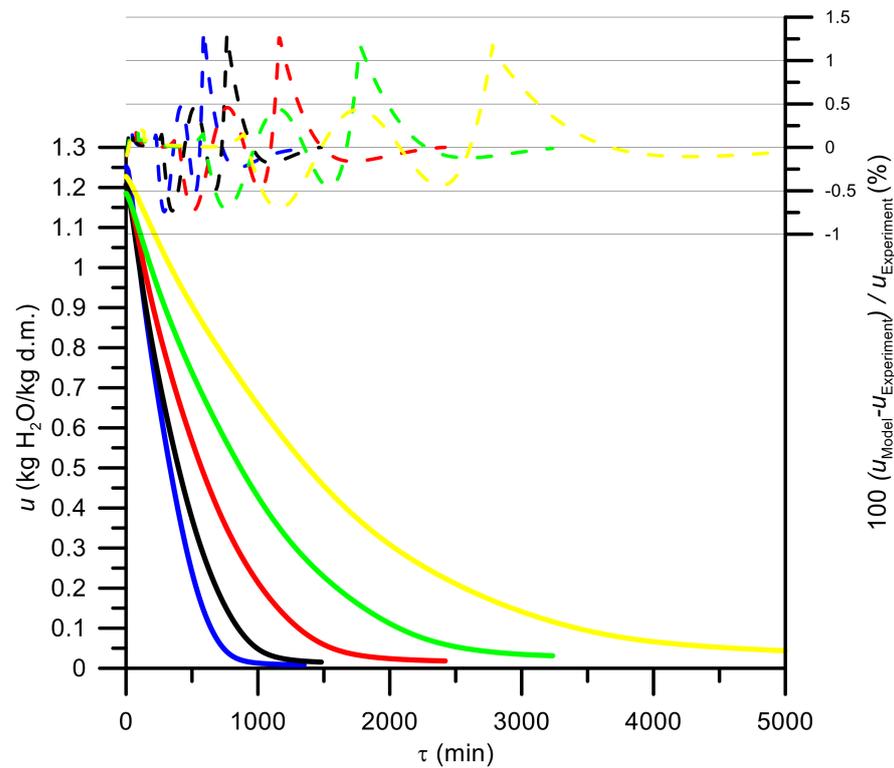


Figure 3. A graph of moisture content versus time of poplar drying at 40 °C (—), 50 °C (—), 60 °C (—), 70 °C (—), and 80 °C (—) and results of mathematical modeling: 40 °C (--- RMSE = 0.0011), 50 °C (--- RMSE = 0.0012), 60 °C (--- RMSE = 0.0011), 70 °C (--- RMSE = 0.0012), and 80 °C (--- RMSE = 0.0011).

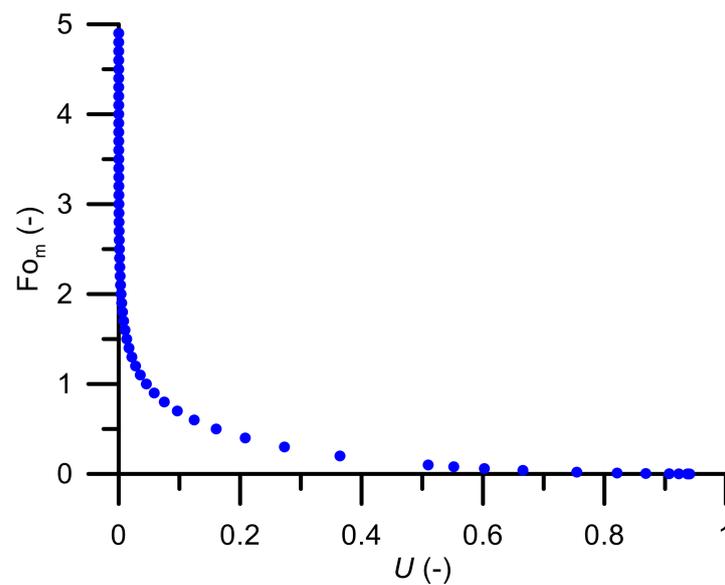


Figure 4. A graph of the value of the Fourier number versus the reduced water content.

As an algorithm for the determination of the mass diffusion coefficient (5) requires a function inverse to the dependence described by Equation (6), such a function, a spline, was obtained by approximation of segments of a set of analytically determined points $[U_i, Fo_{mi}]$. In order to accurately represent the course of changes of coefficient Fo , the above graph (Figure 4) was divided into four parts, which were then approximated with the accuracy of 10^{-5} , within the ranges. Table 2 presents characteristics of the trend line.

Table 2. Intervals and functions approximating Fo .

Interval	Approximating Function $Fo = f - 1(U)$	Coefficient r
$(0; 0.12437>$	$y = -0.405\ln(U) - 0.2508$	$r = 1$
$(0.12437; 0.67>$	$y = 7.8128U^4 - 15.912U^3 + 12.941U^2 - 5.471U + 1.1077$	$r = 1$
$(0.67; 0.9225>$	$y = -0.4395U^3 + 1.4667U^2 - 1.6428U + 0.6133$	$r = 1$
$(0.9225; 1>$	$y = 0.2058U^2 - 0.4392U + 0.231$	$r = 1$

The coefficient r , given in the table, contains information about the model's fitness. When $r = 1$, the fitness of the approximating function is ideal.

Measurements of biomass drying kinetics were used to determine the diffusion coefficient for the examined species of plants. Figure 5 presents an example course of changes in the water diffusion coefficient versus water content for samples dried at a temperature of 60°C .

The internal water diffusion coefficient increases with the decrease of water content until it reaches the water value of approx. 0.025 kg/kg , and then it increases rapidly.

The analysis of the obtained results shows that the diffusion coefficient varies for different plants for the same water content (Figure 5). Graphs of changes of the coefficient a_m for osier, rose, and acacia have similar shapes and ranges while the graph for poplar differs significantly. For the water content above 0.03 kg/kg , the lowest diffusion coefficient was determined for samples of osier, and the highest was determined for poplar, which may be attributed to the lowest density compared with other examined plants.

Figure 6 presents the diffusion coefficient a_m versus water content in energy poplar particles for five drying temperatures.

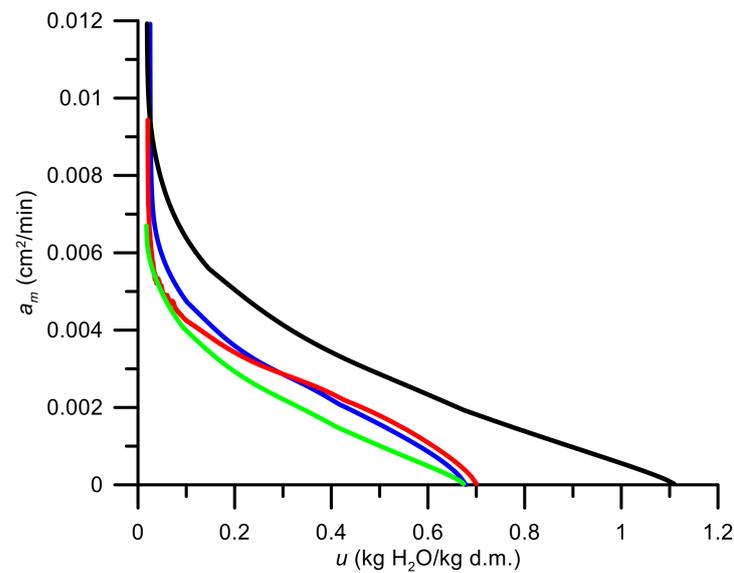


Figure 5. A graph of water diffusion coefficient versus water content in particles of osier (—), rose (—), poplar (—), and acacia (—) dried at 60 °C.

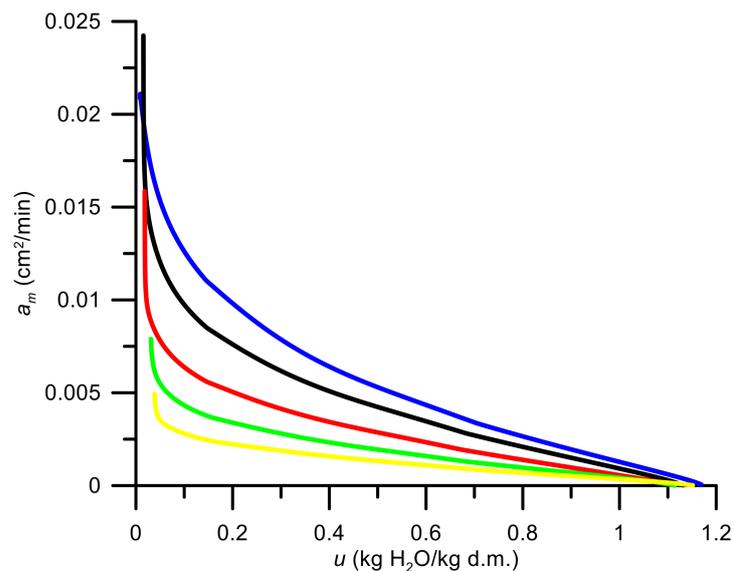


Figure 6. A graph of the diffusion coefficient versus water content in poplar particles drying at 40 °C (—), 50 °C (—), 60 °C (—), 70 °C (—), and 80 °C (—).

As the graphs show, the range of changeability of water content in the examined samples of poplar was the same, while for water content u greater than 0.03 kg/kg, the value of coefficient a_m was greater for higher temperatures of the drying agent, with the greatest value found for the highest drying temperature (80 °C). All curves shown in the figure above have a similar shape for each drying temperature. The value of the diffusion coefficient for water contents lower than approx. 0.03 kg/kg rises sharply at each of the drying temperatures.

The values of the moisture diffusion coefficient for biological materials reported in the literature lie within the range of 10^{-12} to 10^{-8} m^2s^{-1} [49].

Determining the mass diffusion coefficient is very complex, which makes it difficult to develop the most reliable method. Researchers [50,51] have determined this coefficient using traditional sorption methods as well as the inverse modeling method. The results were not satisfactory due to the large error. Difficulties associated with the diffusion can be due to the distribution of the water contained between the cellulosic fibers and the water

contained in the fibers. These relationships were investigated in [52] by use of the pulsed spin-echo gradient (PGSE) method, determining the differences in water diffusion from the fibers and the areas between them. An extension of this method for determining the water diffusion is the spin-echo method (NMR) [53], in which the parameters determine the interconnection of the structural material derived from plants.

The use of various methods to determine the diffusion coefficient allows for a better understanding of its value during the drying process of biological materials.

5. Summary and Conclusions

The determination of the water diffusion coefficient in moist wood biomass during convective drying is an important issue that describes the process of water transport in the material. The coefficient value is determined using an indirect method, which is based on theoretical laws of physics and mathematics. With this method, it is possible to determine the Fourier number, which is extremely important in determining the water diffusion coefficient. In this work, the authors defined a theoretical dependence of the Fourier number on reduced water content in a convectively dried cuboidal solid, based on a simplified theoretical solution of the diffusion equation for such a body. Based on the results of calculations, the following conclusions may be formulated:

- The value of the water diffusion coefficient for all types of examined wood biomass increases with the temperature of the drying agent.
- In all examined cases, within the water content range from initial to approx. 0.03 kg/kg, values of the mass diffusion coefficient increase slightly; after reaching approx. 0.03 kg/kg, they rise sharply.
- Graphs showing changes of the diffusion coefficient versus water content for osier, acacia, and multiflora rose are similar, while the graph for poplar differs.
- At all drying temperatures, values of a_m calculated for osier, acacia, and rose were similar, while for the poplar, those values were considerably higher. This may be attributed to the highest initial water content as well as the smallest density of the material.

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