

*The creation of environmentally friendly protective materials for building structures made of wood could make it possible to influence the processes of stability and the physical-chemical properties at the thermal modification of hornbeam wood over a certain time. That necessitates studying the conditions for investigating phase transformations when the timber is exposed to high temperature, as well as establishing the mechanism of hornbeam wood thermal modification. Given this, a mathematical model of the phase transformation process during the transfer of heat flux to a sample was built. Based on the derived dependences, it was established that when hornbeam wood is exposed to temperature treatment, it undergoes endothermic phase transformations characterized by the heat absorption and change in the color of hornbeam wood. In particular, at a temperature of 200 °C, the temperature in the wood decreases by 5 % due to the chemical changes in the structure of cell wall components (lignin, cellulose, and hemicellulose). It was found that the process of thermal modification is accompanied by the decomposition of hemicellulose and the amorphous part of cellulose, a decrease in moisture absorption, as well as a decrease in the volume of substances that are a medium for the development of fungi. In addition, lignin and the resulting pseudo lignin undergo a process of polymerization and redistribution throughout the cell volume. At the same time, they give the cell walls higher density, hardness, increase hydrophobicity (water repellency), thereby reducing the ability to absorb moisture and swell. It was established that the most effective parameter of phase transformations is the temperature and aging duration. The results of moisture absorption have been given; it has been found that over 6 hours of modified timber exposure, its moisture absorption decreases by more than 10 times, which allows its application at facilities with high humidity*

**Keywords:** *thermally modified timber, modification efficiency, moisture absorption, diffusion, timber moisture resistance*

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# ESTABLISHING REGULARITIES IN THE PROPAGATION OF PHASE TRANSFORMATION FRONT DURING TIMBER THERMAL MODIFICATION

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## 1. Introduction

Representing a building material, timber is widely used in construction and architecture due to its mechanical and operational properties; however, it is destroyed under the influence of atmospheric factors. Improving the operational level of facilities where timber structures are used is possi-

ble through thermal modification. That implies rendering the timber the capability to resist, for example, the effect of moisture, and prevent the destruction of timber thereby decelerating the process of destruction [1].

Given the above, the thermal modification of timber is associated with difficulties related to the use of technological modes such as time, temperature. This is primarily

because the structure and composition of wood differ so that the modification process is not completed while the use of low-heat modified timber leads to destruction. The knowledge of the physical-chemical properties of such substances, their quality indicators, the mechanism that affects materials, would make it possible to select them considering economic indicators, the duration and safety of application, environmental aspects, etc. [2, 3].

Therefore, the development of technological modes of timber modification, studying the thermal-physical transformations, the effect of the structure's features on this process, is an unresolved component of the production of durable building materials from wood. This predetermines the need to establish a mechanism of thermal modification.

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## 2. Literature review and problem statement

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In work [4], it was shown that the thermal modification of timber leads to changes in weight, wettability, and color, thereby increasing its resistance against destruction due to the chemical transformations of timber components. However, issues related to quality control, modeling, and studying the reasons for improving the properties of modified timber remain unresolved. This very approach was used in work [5] where it is shown that one of the best materials for cladding building structures is thermally modified timber because the change in temperature and humidity fields does not affect the intensive destruction. The use of properly selected and correct modification technology could slow down the processes of destruction and environmental consequences. However, the extent and modes of timber modification were not specified.

Extensive use of thermally modified timber [6] has led to the need to create a reliable quality control, which includes control over product deviations within certain limits. That allows a third-party control in the case of certification and regulation of consumer complaints and requirements. However, it is not specified what methods are needed to improve the targeted properties of modified timber during industrial production.

In particular, the authors of [7] devised a new technology of heat treatment in the presence of steam; it is, therefore, a standard hygrothermal treatment. When choosing wood species, one needs to deal with factors such as timber oxidative degradation. An option to overcome the corresponding difficulties may be to determine the reactions that occur through the presence of moisture.

A variant to overcome the relevant problems might be to study changes in the swelling and surface roughness of alder and elmwood after heat treatment at two different temperatures and durations [8]. The results showed that the parameters of swelling and surface roughness differed significantly for two temperatures and two durations of heat treatment. The reason for this may be those objective difficulties that are associated with the value of swelling and surface roughness, which decreased with an increase in the temperature and time of treatment.

A change in the coloration and reflectivity of timber surfaces, caused by artificial weathering obtained in a solar box chamber, which simulates external conditions and subsequent leaching of water, was assessed in [9]. As the weathering time increases, the untreated surfaces of timber samples get darker while the modified samples become

lighter. However, there are unresolved issues related to the possibility of having similar color or, in any case, reducing the starting chromatic difference observed at the beginning of tests for weather-related stability.

Many chemical reactions occur during heat treatment, leading to changes in the components of the primary cell wall of wood and the darkening of the material [10]. Other changes include the resistance of modified timber against fungal decay, making it suitable for use indoors and outdoors as cladding, flooring, floors, garden furniture, and window frames. However, the effectiveness of application was not determined for the most common types of timber, in particular, pine.

Laboratory tests have shown a positive effect of thermal modification on the durability, stability of size, as well as thermal conductivity of timber [11]. The monitoring results showed that the elements and windows made of thermally modified spruce have significantly lower moisture content in timber compared to windows made of unmodified timber. However, the effectiveness of application was not determined when compared with wax and other water repellents.

Article [12] reports a study into the use of artificial aging of timber, which plays an important role in assessing the results of work by reducing time compared to natural weathering. The approach is to protect the surface by using various types of commercially-available agents such as solvents contained in water, with large content of solids, powder coatings. However, the effectiveness of thermal modification of timber, its resistance to artificial aging is not shown.

One of the approaches to improve the durability of timber is a set of processes that provide the treated material with a better capability to cope with the damage caused by the external environment, by increasing the duration of treatment [13]. It was established that this process is also performed to enhance the physical, mechanical, or aesthetic properties of timber, and makes it possible to obtain products that are not harmful to users and the environment, similar to natural wood [14]. However, the mechanism of timber degradation caused by operation and its impact on destruction rates were not specified.

Thus, the scientific literature has revealed that the thermal modification of timber could reduce the destruction of a building structure and expand the scope of timber application. The above allows us to assert that it is expedient to conduct a study to determine the parameters that ensure the resistance against destruction, as well as define the mechanism affecting timber transformation exposed to thermal modification. Establishing the operational mechanism involved in the thermal modification of timber to render its resistance to moisture absorption has predetermined our research in this area.

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## 3. The aim and objectives of the study

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This work aims to establish the parameters of phase transformations during timber thermal modification. That could improve the technology of manufacturing such products and expand the scope of their application.

To achieve the set aim, the following tasks have been solved:

- to model the process of phase transformations' front propagation when timber is exposed to thermal influence due to high temperature;

– to establish features in the decrease of moisture absorption in order to confirm the termination of phase transformations during timber thermal modification.

**4. Materials and methods to study the process of timber thermal modification**

**4.1. The examined materials and equipment used in the experiment**

Hornbeam timber, which belongs to hardwood, is used for the manufacture of floors in rooms with high humidity (swimming pools, saunas); to reduce the rate of water absorption, it is advisable to conduct thermal modification.

Our research involved samples of raw hornbeam wood of 25×20×20 mm (Fig. 1).

The thermal modification of timber was conducted at a temperature of 200 °C over 1–6 hours with a 1-hour interval.



Fig. 1. A model sample of timber

To establish the degree of timber water absorption, the samples of thermally modified timber were used.

**4.2. Procedure for establishing the indicators of samples' properties**

Since the phase transformations during timber thermal modification occur at the intercellular level, it is impossible to install a measuring device inside the timber to measure the temperature without damaging its structure; and the destruction of timber could lead to erroneous results. Our study, aimed at modeling the process of propagation of the phase transformations' front during timber thermal modification, was performed using the basic provisions from mathematical physics [15].

We determined the degree of moisture absorption by timber according to the working procedure whose essence was to experimentally establish the amount of the water absorbed by a sample at its exposure to 100 % moisture. To find the values of the amount of water absorbed by timber, specialized equipment was designed and manufactured (Fig. 2).

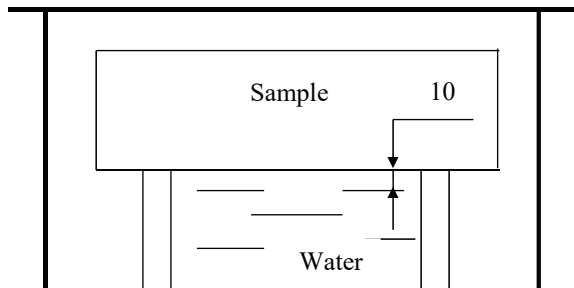


Fig. 2. Device for testing water absorption by timber

A tested sample was fixed in a special cuvette so that it was in a humid environment above the water. After a cer-

tain time, the sample was weighed on scales; the amount of water absorbed was determined.

Based on the measured values, we registered the changes and determined the efficiency of timber thermal modification.

**5. Modelling the process of timber thermal conductivity at its thermal modification**

As a result of the thermal treatment of timber, under the action of heat flow, the direction of the decomposition of hemicellulose, the redistribution of lignin, etc. changes, towards the formation of volatile products and carbonized residue. This process is characterized by heat absorption during phase transformations and discoloration of hornbeam wood. Considering the above, the issue arises about studying the phase transformations of timber exposed to heat.

It should be noted that determining the thermal-physical characteristics of thermally modified timber is associated with some obstacles, namely the measurement of temperature in a timber layer that changes over time.

To establish the temperature of phase transformations within thermally modified timber, a method has been proposed to solve the problem of thermal conductivity for a plate with the thermal-physical properties that depend on temperature. Over the initial time, a time-constant heat flux  $q_0$  is applied to the surface of a timber sample. That is, it is instantly heated to a temperature that is kept constant throughout the heating process; the temperature distribution passes to the center of the sample (Fig. 3).

Three regions were considered (Fig. 3):

- 1 – external environment,  $x < 0$ ;
- 2 – phase transition zone,  $0 < x \leq Z$  ( $Z$  is the hemicellulose conversion coordinate, m);
- 3 – timber (material of the sample of hard substance) ( $R - Z$ ), m.

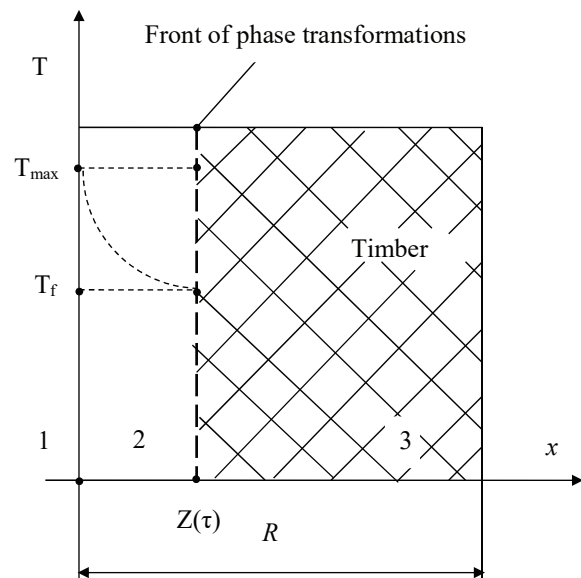


Fig. 3. The scheme of the process of propagation of the phase transformations' front during timber thermal modification: 1 – external environment; 2 – phase transition zone; 3 – timber

The differential equation of heat transfer in timber takes the following form:

$$\frac{\partial^2 T(x, \tau)}{\partial x^2} - \frac{1}{a} \frac{\partial T(x, \tau)}{\partial \tau} = 0, \quad (x > Z(t); t > 0), \quad (1)$$

at the initial and boundary conditions

$$T(x, t)|_{t=0} = 0, \quad x > 0; \quad (2)$$

$$T(x, t)|_{x=Z(t)} = \phi(t), \quad t > 0; \quad (3)$$

$$T(x, t)|_{x=\infty} \rightarrow +\infty, \quad x \geq 0, \quad (4)$$

where  $a$  is the coefficients of temperature timber conductivity,  $\text{m}^2 \cdot \text{s}^{-1}$ ;  $Z(t)$  is the coordinate of timber phase transformations;  $\phi(t)$  is the rate of timber phase transition changes;  $T$  is the temperature,  $^{\circ}\text{C}$ ;  $x$  is a coordinate,  $\text{m}$ .

A solution to the problem can be represented in the following form:

$$T(x, t) = \frac{\sqrt{a}}{2\sqrt{\pi}} \int_0^t \psi(\tau) \frac{1}{\sqrt{t-\tau}} \cdot e^{-\frac{(x-Z\tau)^2}{4a(t-\tau)}} d\tau. \quad (5)$$

The unknown function  $\psi(\tau)$  can be found from a condition at the boundary  $x = Z \cdot t$ :

$$\phi(t) e^{\frac{Z^2 t}{4a}} = \frac{\sqrt{a}}{2\sqrt{\pi}} \int_0^t \psi(\tau) \frac{1}{\sqrt{t-\tau}} \cdot e^{-\frac{Z^2 t}{4a}} d\tau. \quad (6)$$

However, the solution to equation (5) is cumbersome, so, by using a Laplace transform from [16], we can write down:

$$\begin{aligned} \bar{T}(x, p) &= \frac{\sqrt{a}}{2\sqrt{\pi}} \int_0^{\infty} e^{-pt} \int_0^t \frac{\psi(\tau)}{\sqrt{t-\tau}} \cdot e^{-\frac{(x-Z\tau)^2}{4a(t-\tau)}} d\tau \cdot dt = \\ &= \frac{\sqrt{a}}{2\sqrt{\pi}} \int_0^{\infty} \psi(\tau) \int_{\tau}^{\infty} \frac{1}{\sqrt{t-\tau}} \cdot e^{-pt - \frac{(x-Z\tau)^2}{4a(t-\tau)}} d\tau \cdot dt. \end{aligned} \quad (7)$$

The original of this equation at any point  $x$  at moment  $t$  is:

$$T(x, 0) = \frac{1}{2\sqrt{a\pi}} \int_0^1 \frac{x-Z(t)}{(t-\tau)^{3/2}} \cdot e^{-\frac{(x-Z(t))^2}{4a(t-\tau)}} \cdot \phi(t) \cdot d\tau. \quad (8)$$

The solution to equation (8), taking into account the initial and boundary conditions in the region of L-transforms, takes the following form:

$$\bar{T}(x, p) = \bar{\phi} \left( p - \frac{Z}{\sqrt{a}} \cdot \sqrt{p} \right) \cdot \frac{1}{\sqrt{p}} \left( \sqrt{p} - \frac{Z}{2\sqrt{a}} \right) \cdot e^{-\frac{x}{\sqrt{a}} \sqrt{p}}. \quad (9)$$

Thus, the argument of the action function  $\phi$  depends not on the parameter  $p$  but the difference:

$$p - \frac{Z}{\sqrt{a}} \cdot \sqrt{p}. \quad (10)$$

Since  $\phi(t)$  is given by the following function:

$$\phi(t) = T_0 \cdot Z(t). \quad (11)$$

Then

$$\phi(p) = \frac{T_0}{p}, \quad (12)$$

$$\bar{\phi} \left( p - \frac{Z}{\sqrt{a}} \cdot \sqrt{p} \right) = \frac{T_0}{p - \frac{Z}{\sqrt{a}} \cdot \sqrt{p}}. \quad (13)$$

Considering (11) to (13), let us write down equation (9) in the following form:

$$\begin{aligned} \bar{T}(x, p) &= \frac{T_0}{p} \frac{\sqrt{p}}{\sqrt{p} - \frac{Z}{\sqrt{a}}} \cdot e^{-\frac{x}{\sqrt{a}} \sqrt{p}} - \\ &- \frac{1}{2} \frac{T_0}{p} \frac{Z}{\sqrt{a}} \cdot \frac{1}{\sqrt{p} - \frac{Z}{\sqrt{a}}} \cdot e^{-\frac{x}{\sqrt{a}} \sqrt{p}}. \end{aligned} \quad (14)$$

Using a ratio from [15], we can write

$$\frac{e^{-\frac{x}{\sqrt{a}} \sqrt{p}}}{\sqrt{p} \left( \sqrt{p} - \frac{Z}{\sqrt{a}} \right)} \rightarrow e^{-\frac{xZ}{a} \left( \frac{Z}{\sqrt{a}} \right)^2 t} \cdot \operatorname{erfc} \left( \frac{x}{\sqrt{a}} - \frac{Z}{\sqrt{a}} \sqrt{t} \right); \quad (15)$$

$$\begin{aligned} \frac{\frac{Z}{\sqrt{a}}}{\sqrt{p} \left( \sqrt{p} - \frac{Z}{\sqrt{a}} \right)} e^{-\frac{x}{\sqrt{a}} \sqrt{p}} &\rightarrow \operatorname{erfc} \left( \frac{x}{\sqrt{a}} - \frac{Z}{\sqrt{a}} \sqrt{t} \right) - \\ - e^{-\frac{xZ}{a} \left( \frac{Z}{\sqrt{a}} \right)^2 t} \cdot \operatorname{erfc} \left( \frac{x}{\sqrt{a}} - \frac{Z}{\sqrt{a}} \sqrt{t} \right). \end{aligned} \quad (16)$$

Considering (11), (12), let us write down the solution to the problem in a time domain:

$$T(x, t) = T_0 \left[ \frac{3}{2} e^{-\frac{Z(x-Zt)}{a}} \operatorname{erfc} \left( \frac{x-Zt}{2\sqrt{at}} \right) - \frac{1}{2} \operatorname{erfc} \frac{x}{2\sqrt{at}} \right]. \quad (17)$$

At  $Z=0$ , when a timber phase transition change is finished, the solution will take the following form:

$$T(x, t) = T_0 \cdot \operatorname{erfc} \frac{x}{2\sqrt{at}}. \quad (18)$$

The derived equation allows calculating the temperature of phase transformations during timber thermal modification based on the experimental values of temperature, the geometric dimensions of a timber sample, and timber thermal conductivity.

Fig. 4 shows the calculation of temperature in a timber sample depending on the time of phase transformations, defined from equation (18) and by applying data from [15], namely the value of the thermal conductivity coefficient of timber at a temperature of about  $200^{\circ}\text{C}$ , which is  $18 \cdot 10^{-6} \text{ m}^2/\text{s}$ .

When timber is exposed to thermal treatment, it undergoes endothermic phase transformations, which are characterized by heat absorption and timber discoloration. In particular, at  $200^{\circ}\text{C}$ , the temperature in the timber, due to the chemical changes in the structure of the components of the cell wall (lignin, cellulose, and hemicellulose), was  $200^{\circ}\text{C}$ ; according to Fig. 4, it was reduced, due to phase transformations, by 3%.

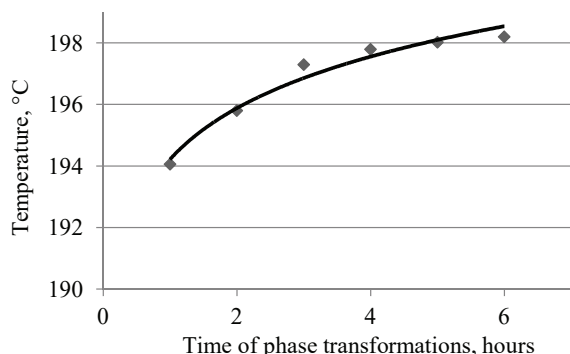


Fig. 4. The dependence of a timber sample's temperature on the time of phase transformations (points correspond to the values calculated from equation (18), averaged by a trend's line)

Thus, in the process of thermal modification, hemicelluloses and the amorphous part of cellulose decompose, moisture absorption decreases, and the number of substances that are the medium for the development of fungi decreases. In addition, lignin and the formed pseudo lignin undergo a process of polymerization and redistribution throughout the volume of the cell, which gives the cell walls greater density, hardness. As a result, hydrophobicity (water repellency) increases, and the capacity to absorb moisture and swell decreases. Polymerized lignin fills the inner cavity of the cell thereby forming a closed porous structure with a low capability to bind water.

It is established (Fig. 4) that the most effective parameter of phase transformations is temperature and exposure time.

Thus, we have derived the dependence that is necessary to predict the end time of phase transitions during timber thermal modification, which makes it possible to directly calculate the movement of timber phase transformations depending on the time of temperature impact.

### 6. Results of studying the features related to the decreased water absorption by the thermally modified timber

To establish the thermal-physical characteristics of thermally modified timber, we performed a study into the modification under the impact of a heating device at 200 °C over a certain time of modification (Fig. 5).

It should be noted that thermal modification should be performed at temperatures above 200 °C; however, it is necessary to take into consideration the ignition temperature of timber, which is from 215 °C to 225 °C, or carry out the process in a protective environment. Therefore, in a given case, taking into consideration safety requirements, the thermal modification was conducted at 200 °C.

As can be seen from Fig. 5, during the modification the hornbeam timber changed color from light to dark depending on the exposure time, respectively, and underwent phase transformations of the components and, accordingly, acquired other properties, in particular moisture absorption.

The results of moisture absorption by timber are given in Table 1.

Our results (Table 1) show that when the timber is thermally modified for 6 hours, the moisture absorption is reduced by more than 2.4 times, which allows it to be used at facilities with high humidity.

Fig. 6 shows the moisture absorption of hornbeam timber depending on the exposure time in a humid environment.

As can be seen from Fig. 6, the largest amount of absorbed moisture was recorded for the untreated hornbeam timber. Thermal modification gradually reduces the ability of the timber to absorb water.

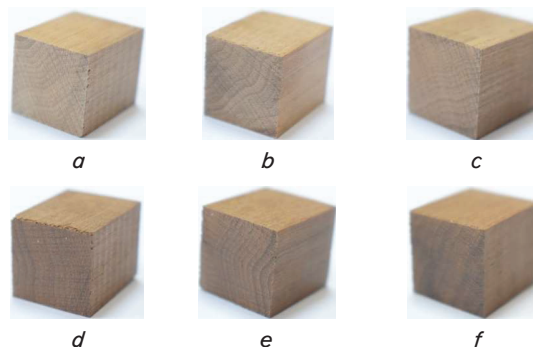


Fig. 5. Model samples of the hornbeam timber thermally modified at a temperature of 200 °C during: a – 1 hour; b – 2 hours; c – 3 hours; d – 4 hours; e – 5 hours; f – 6 hours

Table 1

Results of the experimentally determined timber water absorption

Sample No.	Gain in the absorbed water, g							
	Time of exposure, day							
	0	1	2	6	8	12	21	26
0	6.80	8.1	8.35	8.62	8.73	8.75	8.76	8.76
1	7.55	8.03	8.31	8.59	8.57	8.63	8.70	8.72
2	7.34	7.73	7.99	8.3	8.38	8.33	8.38	8.48
3	7.38	7.67	7.97	8.24	8.30	8.34	8.44	8.44
4	7.11	7.32	7.69	7.9	8.00	8.07	8.11	8.12
5	7.26	7.46	7.74	7.89	7.93	7.99	7.99	8
6	7.19	7.39	7.6	7.8	7.79	7.86	7.86	7.86

### 7. Discussion of results of studying the process of propagation of the phase transformations' front at timber thermal modification

During the thermal modification of timber, as indicated by the research results, both theoretical equations (18) and Fig. 4 and the experimental study illustrated by Fig. 5, there is a natural process of the chemical transformations of hornbeam wood components. Namely, color changes under the impact of temperature and, accordingly, changes in structure, which, in turn, could lead to a decrease in moisture absorption. Phase transformations in the structure of wood under a temperature impact are characterized by an endothermic effect, which decreases over time. The thermally modified timber, at the same exposure temperature but over a longer time, is characterized by lower moisture absorption. Such a mechanism of thermal timber modification is probably a factor in regulating the extent of the formation of weather-resistant material. This agrees with the data known from [5, 8], the authors of which also link the effectiveness of protection against moisture during the thermal modification of hornbeam wood. In contrast to the results reported in [4, 7], our findings on the process of moisture absorption by thermally modified hornbeam timber and changes in the moisture insulating properties of hornbeam timber allow us to state the following:

– the main regulator of the process of moisture absorption is not only the formation of the coating layer but also the chemical transformations of the components of hornbeam wood, which, under the impact of temperature and humidity fields, provide for the resistance against moisture;

– a significant impact on the process of protection of thermally modified hornbeam timber from moisture fluctuations is exerted towards the formation of water-resistant capillary porous elements throughout the volume and at the surface of the natural material.

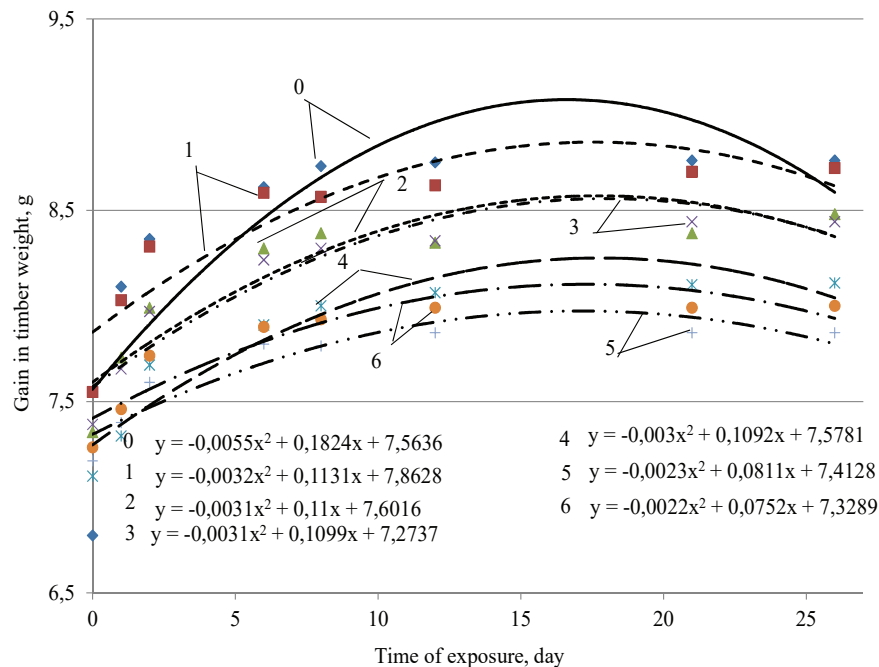


Fig. 6. The dependence of the amount of absorbed water on the exposure time of hornbeam timber

Such conclusions can be considered reasonable from a practical point of view because they allow for a substantiated approach to determining the necessary technology for the thermal modification of hornbeam timber. From a theoretical point of view, they allow us to argue on determining the mechanism of phase transformation processes, which is a certain advantage of this study. The results of determining the moisture absorption during thermal modification (Table 1) indicate the ambiguous effect exerted on the nature of the change in humidity by the modified hornbeam timber. In particular, this implies the availability of data sufficient for the quality process of the inhibition of moisture diffusion, and the detection, on its basis, of the time when a decline in moisture resistance begins. This

detection could make it possible to study the transformation of hornbeam timber, which moves in the direction of increased resistance to destruction, and to identify those variables that significantly affect the onset of the transformation of this process.

The results of the current study are limited to the use of the thermally modified hornbeam timber; it is necessary to refine the data when applying other types of wood.

## 9. Conclusions

1. We have modeled the process of propagation of the phase transformations' front, and established the dependence of temperature on the time of hornbeam timber exposure, as well as derived the estimation dependences that could help find a change in the temperature dynamics during phase transformations. At a temperature exposure, endothermic phase transformations take place in hornbeam timber, which are characterized by heat absorption and discoloration of hornbeam wood. In particular, at a temperature of 200 °C, the temperature in hornbeam timber, due to the chemical changes in the structure of the cell wall (lignin, cellulose, and hemicellulose), is reduced by up to 3%.

2. Special features in the inhibition of the process of moisture advancement to the thermally modified hornbeam timber imply several aspects, namely, the formation of waterproof components, as well as capillary porous elements, which are characterized by the

formation, at the hornbeam timber surface, of a waterproof layer. Thermal modification of hornbeam timber reduces moisture absorption by more than 2.4 times within 6 hours, which allows it to be used at facilities with high humidity.

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