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Graphene Oxide as a Factor Modifying the Properties of Wood

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Abstract: This work carried out research to determine the possibilities of using graphene oxide to provide wood with new functional features. With the saturation parameters used and working liquid with a concentration of 0.004% graphene oxide, the retention of the nanomaterial in wood was 0.25 kg/m^3. The presence of graphene oxide increased the crystallinity of the wood to 64% (compared with 57% for unmodified wood). The TG/DTG spectra of wood impregnated with graphene oxide and the control wood indicated that the initial weight loss of the samples observed at a temperature of 100 °C was similar and amounted to less than 4%. A second mass loss was observed in a temperature range of 270 to 380 °C. The mass loss in this temperature range reached 70% and was similar in the test and control samples. Wood modified with graphene oxide showed increased thermal stability in a temperature range of 360 to 660 °C compared with native wood. Given the results obtained, there were no statistically significant differences in the water absorption of modified or control wood. The presence of low concentrations of graphene oxide in the culture medium did not inhibit the growth of the fungus *Trichoderma viride*; however, a decrease in the growth activity of mycelial hyphae was observed with an increasing concentration of nanomaterial in the medium. It has been reported that graphene oxide, as a stress factor, initiates changes at the cellular level, characterized by the formation of structures called chlamydospores by the body.

Keywords: graphene oxide; wood properties; crystallinity; TG/DTG; mold fungi

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1. Introduction

The search for methods to expand the functionality and increase the durability of wood is one of the most important trends in the development of wood materials science. Wood is an excellent utility material, but its durability depends on the conditions of use. Improper climatic and humidity conditions lead to the loss of the desired physical and mechanical properties of wood and often even to its permanent degradation. The most important factor influencing structural changes in wood is microorganisms, the development of which is possible when its humidity is appropriately increased [1]. Both basidiomycete fungi and molds develop on wood with increased humidity. The chemical modification of wood leads to changes in its properties and susceptibility to biocorrosion; however, there are still no methods or means for the fully effective and long-term protection of wood against changing climatic factors [2,3].

In recent years, there has been a trend of nanotechnology entering the field of woodworking. The use of graphene oxide to increase the thermal conductivity of wood was the subject of research by Zhang et al. [4]. The authors of that publication showed that graphene and its derivatives significantly increase the thermal conductivity of veneers,

thus expanding the scope of their potential applications. Interesting research in this area was conducted by He et al. [5], proving that reduced graphene oxide provides fir wood with decent anisotropic conduction without reducing its strength parameters.

In another study by Xu et al. [6], the use of graphene oxide as an addition to polyurethane paints used for finishing furniture resulted in a significant increase in the hardness of the coating and scratch resistance. In turn, Han et al. [7] focused on analyzing the impact of graphene oxide on changes in the flammability of wood, indicating that it may play an important role in fire protection.

Recently published research results indicate that graphene oxide added to phenol-formaldehyde (PF) resin significantly improves the compressive strength, elastic modulus, and moisture resistance of plywood panels [8]. The same authors also demonstrated that graphene oxide is very effective at capturing formaldehyde, which makes the production of wood-based materials based on PF resins more environmentally friendly.

Graphene oxide (GO) is a two-dimensional material with single-layer carbon atoms and numerous functional groups providing it with high reactivity. Numerous scientific projects have proven that this nanomaterial increases mechanical strength and contributes to an increase in the hydrophobicity of materials [6,9,10]. From the practical point of view of maintaining the durability of wood, increasing its strength and resistance to moisture are key elements in maintaining its durability.

The use of nanomaterials as superhydrophobic agents for wood technology applications was detailed in research conducted by Łukawski et al. [11]. Researchers have analyzed the effect of graphene and nanotubes on wood fibers and particles, leading to a change in the nature of these particles from naturally hydrophilic to superhydrophobic. The obtained results indicate new possibilities for the chemical modification of wood toward changes in the degree of wettability of the wood surface and moisture absorption [12]. These studies provide grounds for stating that nanomaterials may constitute prospective solutions in the development of methods and techniques for protecting wood against biological degradation [13].

It should be additionally noted that the use of nanotechnology as a method of modifying wood and providing it with new features and properties is an interesting issue, for example, because of the lack of detailed knowledge about changes in the molecular structure; functionality; and, most importantly, safety of using wood–nanomaterial composites [14].

Wood protection strategies related to the development of methods and means of protection are increasingly turning to nanomaterials. The search for new solutions to protect wood against biocorrosion prompted the authors of this publication to conduct research on the potential possibilities of using graphene oxide in order to provide wood with new functional features. However, it should be mentioned that fungicide effectiveness was not the primary concern. Any chemical modification may lead to changes in the physical and mechanical parameters of wood. When analyzing each new active substance as a potential component of wood protection product formulations, it should be ensured that it does not reduce the physical and strength parameters of materials, does not contribute to increased flammability, and is not harmful to the environment and users of impregnated wood. Therefore, in the presented study, the authors of this research set themselves the following research goals: (1) to determine whether the process of impregnating pinewood with a water dispersion of graphene oxide in a small vacuum leads to the modification of the wood, which may result in new material properties; (2) to determine whether graphene oxide in low concentrations can constitute a potential–special fungicide. There are many works in the literature devoted to the assessment of the fungicide properties of graphene oxide, but it is difficult to find research devoted to the issue of wood protection. Wood protection is a very broad issue that should be analyzed in many aspects, not only in terms of the biocidal activity of the substance itself but also its impact on the impregnated material and, in particular, its impact on the environment.

2. Materials and Methods

2.1. Preparation of Research Material

Pinewood samples (sapwood) (*Pinus sylvestris* L.) with dimensions of 40 × 40 × 4 mm were used for the tests (Figure 1). The density of the wood at 12% humidity was 400 kg/m^3. Graphene oxide dispersion (Advanced Graphene Products S.A., Zielona Góra, Poland) was used to impregnate the wood. The diameters of the graphene oxide flakes were 500–1000 nm. A 0.004% suspension of graphene oxide in distilled water was exposed to ultrasound PS-40A (CNCWorld Group Sp. z o.o., Jedlnia Letnisko, Poland). Graphene oxide (GO) in the form of an aqueous dispersion was used to prepare a solution with which to impregnate the wood samples. In order to achieve homogeneity in the solution, the wood was ultrasonically treated before impregnation. Impregnation of the samples was carried out under vacuum in a vacuum dryer, model 1445-2 (Sheldon Manufacturing Inc., Cornelius, OR, USA) with a pump—model V-700 (BUCHI Labortechnik AG, Flawi, Switzerland). Impregnation parameters: vacuum, 190 mbar; time, 30 min; and temperature, 28 °C. After this time, the pressures in the autoclave were brought to atmospheric pressure, and the samples remained in solution for a further 30 min. The samples were then removed from the graphene oxide dispersion. Based on the initial weight of the samples and their weight after impregnation, the retention of graphene oxide in the samples was determined.

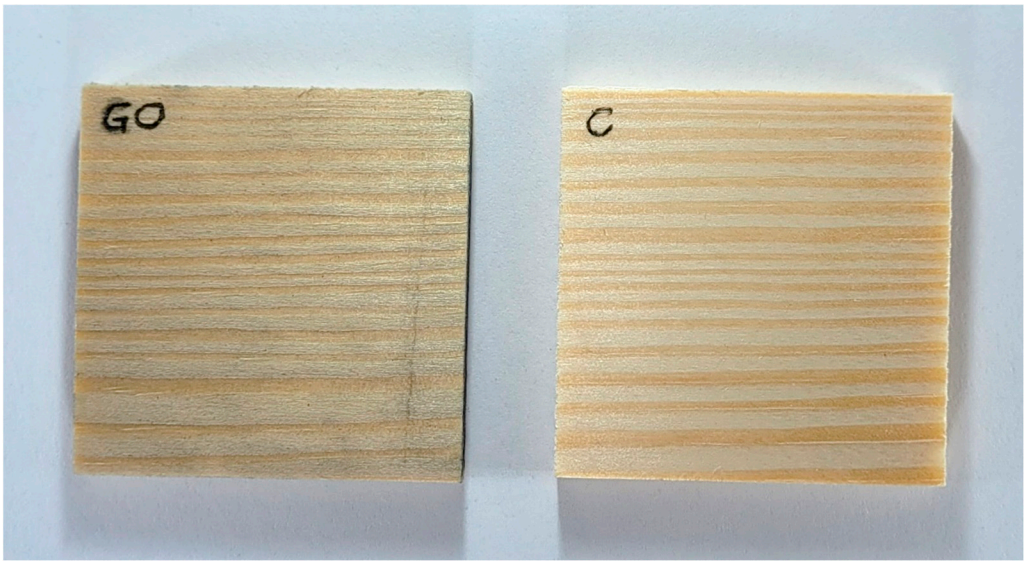


Figure 1. Samples: GO—wood-GO; C—control.

2.2. Characterization of Quality Parameters

2.2.1. XRD Analysis

X-ray diffraction (XRD) was measured by using a SmartLab X-ray diffractometer (Rigaku Corporation, Akishima, Japan) with a Ni-filtered Cu Kα (1.5418 Å) radiation source operated at a voltage of 40 kV and 30 mA of anode excitation. The X-ray diffraction pattern was recorded for the angles at a range of 2Θ = 5–30° in a step of 0.04°/3 s. The crystallinity (X_c) of the materials was calculated as the ratio of the total area under the resolved crystalline peaks to the sum of the area under the crystalline peaks and the amorphous curve. The following formula was used (1):

$$X_C = \frac{\Sigma A_C}{\Sigma A_C + A_A}\%, \quad (1)$$

where
A_C—total area under the resolved crystalline peaks;
A_A—total area of amorphous phase.

2.2.2. TG/DTG Analysis

Thermogravimetric (TG) measurements and the derivative thermogravimetric (DTG) measurements were carried out using a Netzsch TG 209F3 apparatus (Netzsch-Gruppe, Selb, Germany). All the sampling was conducted in a temperature range of 40–700 °C at a heating rate of 10 K min^{-1} in a nitrogen atmosphere. The mass change was recorded as a function of temperature. The peak temperature of maximum thermal degradation (DTG peak temperature) was designated as the maximum temperature obtained from the differentiation of the mass change as a function of temperature.

2.2.3. SEM Analysis

After impregnating white pinewood samples, their morphology was evaluated through scanning electron microscopy (SEM) using a SEI QUANTA electron microscope (FEI Company, Hillsboro, OR, USA). The samples exposed to graphene oxide were cut into 5 µm thick sections using a cryostat (CM 1900, Leica, Wetzlar, Germany). These sections were attached to microscope slides coated with poly-l-lysine and then dehydrated in increasing concentrations of hexylene glycol (Sigma-Aldrich, St. Louis, MO, USA). A Polaron CPD 7501 critical point dryer (Quorum Technologies, Laughton, UK) was used for drying. Finally, the samples were placed on aluminum SEM stubs and next inspected via SEM at 1 keV.

2.2.4. Assessment of Water Absorption

A study of the absorption of pinewood impregnated with graphene oxide was carried out based on a methodology according to [15], modified by extending the test time. Before starting the absorption test, the impregnated wood samples were dried to a constant weight (W = 0%) using a drying machine (JP Selecta, Barcelona, Spain). The impregnated wood samples were then placed in glass vessels with graphene oxide solution and loaded with glass weights. The weight of the samples was measured after 0, 5, 10, 15, 25, 35, 45, 60, 75, 90, 120, 180, 1140 (24 h), 2880 (48 h), 4320 (72 h), and 5760 (96 h) minutes. The weight of the samples was taken using a precision balance model, model WPS210/C/2 (Radwag, Radom, Poland), with an accuracy of 0.001 g. Analysis of variance using the Snedecor statistic was used to statistically verify the obtained results. Statistical inference was performed for a significance level of $\alpha = 0.05$. If the null hypothesis was rejected, the Tukey test was performed. The statistical hypothesis was as follows: H0: absorption wood-GO = absorption wood–control; H1: the absorptions tested are different.

2.3. Characterization of Biological Parameters

2.3.1. Growth of Fungi on Wood

Wood samples impregnated with graphene oxide and the control were placed on Petri shackles overgrown with the mycelium of Trichoderma viride Pers., strain A-102. The samples were placed on sterile glass supports to prevent the direct impact of the medium on the wood. Wood samples for testing were sterilized in a steam autoclave model: Classic Standard 210001 (Prestige Medical, Blackburn, UK).

The attack of mold fungi on wood is superficial, especially in the first stage of this attack; therefore, the influence of graphene oxide on the fungi was assessed using the superficial fouling methodology. The fungal growth of the wood samples was determined on the basis of high-resolution photographic images taken daily for each tested sample. The growth of fungi on the sample was determined as the percentage of the sample area covered with mycelium compared with the total area of the tested sample. The percentage of growth on the wood samples was determined to the nearest 5% using the ImageJ2 image analysis software (Fiji v.1.52i).

2.3.2. Growth of Fungi on Sabouraud Dextrose Agar Medium

The effect of graphene oxide on the growth of mold fungi was determined on Sabouraud Dextrose Agar (OXOID Ltd., Basingstoke, UK). *Trichoderma viride* Pers., strain A-102, from a collection of pure cultures from the Department of Wood Science and Protection of the

Warsaw University of Life Sciences (SGGW), was used in this research. Subsequently, 0.004% dispersion graphene oxide was added to a sterile Petri dish in the following amounts: 0.01; 0.05; 0.1; 0.25; 0.5; and 1.0 mL/10 mL culture medium. Mycelium inoculum was introduced centrally onto the solidified medium. The size of the inoculum was 5–6 mm. The culture was carried out in a thermal incubator model, Thermolyne Type 42000 (ThermoFisher Scientific, Waltham, MA, USA), under temperature and relative air humidity conditions of 26 ± 2 °C and 63 ± 2%, respectively. The assessment of fungicide properties on the culture medium was carried out by measuring the diameter of mycelium growth in two perpendicular directions. Measurements were performed at 48 h intervals. The tests were completed on the day when the substrate was completely covered in the controls. To verify the statistical analysis, analysis of variance using the Snedecor statistic was used. Statistical inference was performed for a significance level of $\alpha = 0.05$. If the null hypothesis was rejected, the Tukey test was performed. The statistical hypothesis was as follows: H0: Ø 0.01 = Ø 05 = Ø 0.1 = Ø 0.25 = Ø 0.5 = Ø 1.0 = Ø control; H1: There are at least two means that differ significantly.

2.3.3. The Effect of Graphene Oxide on Mold Fungus Cells

In order to visualize the interaction of graphene oxide with fungal cells, fungal growth tests were carried out on a microbiological medium with the addition of graphene oxide labeled with fluorescein isothiocyanate.

In total, 0.1 mg of fluorescein isothiocyanate (FITC; F143, Thermo Fischer Scientific, Waltham, MA, USA) was used to label the GO, and it was incubated overnight at 4 °C, protected from light. GO conjugated with FITC was added to the Petri dish at an amount equal to 0.1 mL/10 mL of medium. After the medium solidified, the mycelium was inoculated in a similar manner as described in previous studies. On the 7th day of culturing, the influence of GO on the morphology of the mycelium was analyzed.

The interaction between GO conjugated with FITC and mycelial hyphae was recorded with a confocal microscope (IX 81 FV-1000, Olympus Corporation, Tokyo, Japan). Image analysis in confocal mode, Nomarski interference contrast, and cell counting were performed using the FVIO-ASW ver. 1.7c software (Olympus Corporation, Tokyo, Japan). Three-dimensional images were assembled from 30 optical sections.

3. Results

The retention of graphene oxide in wood is shown in Table 1. From the comparison of liquid retention between the modified and control wood, it can be seen that GO does not block the flow of water in the wood. With the saturation parameters used and the working liquid with a concentration of 0.004% graphene oxide, the retention of the nanomaterial in wood was 0.25 kg/m^3. The obtained retention is much lower than the recommended retention of fungicide substances in commercial wood impregnation agents. The use of such a low retention rate was dictated by the initial assumptions of the research in order to introduce the smallest possible number of substances into the wood and to observe changes in the material properties that resulted from the modification.

Table 1. Retention of graphene oxide-treated wood samples.

Samples	Solution Retention		Graphene Oxide Retention	
	Average Content	SD	Average Content	SD
	kg/m^3			
wood-GO	636.22	26.32	0.25	0.01
control	637.75	8.01	-	-

SD—standard deviation.

3.1. Characterization of Structural Parameters

3.1.1. XRD Analysis and the Degree of Crystallinity

The XRD analysis carried out illustrates micro-changes in the structure of pinewood as a result of the impregnation process with a water dispersion of graphene oxide (Figure 2). The profile of the curves is characteristic of wood containing cellulose [16,17]. Three characteristic peaks can be identified at 2θ diffraction angles of 14.88°, 16.68°, and 22.6°, which correspond to lattice planes with Miller indices amounting to (010), (110), and (200). The location of the maxima at these 2θ angles indicates the presence of polymorphic cellulose I [18].

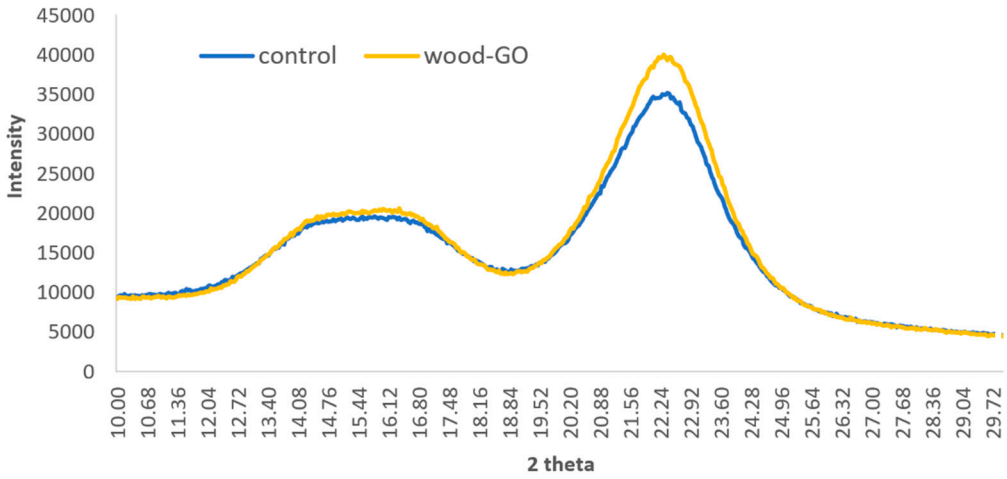


Figure 2. XRD patterns of wood and wood-GO.

The degree of crystallinity of pinewood is 57%, which is consistent with the results of analyses by other authors [19]. The presence of graphene oxide increases the crystallinity of the material to 64% (Table 2). The increase in cellulose crystallinity may be related to the formation of bonds between GO and the -OH groups of cellulose in the amorphous fraction, which may increase intermolecular interactions and, consequently, increase the molecular order [20]. This type of assumption was confirmed by research conducted by Chen et al. [21], which indicated that GO nanosheets form covalent bonds with the aluminophosphate used in the wood-based panel industry as a fiber-gluing component. In another publication [22], the authors indicate that the increase in crystallinity may also be associated with the possibility of the destruction of polysaccharide chains present in the amorphous areas of cellulose.

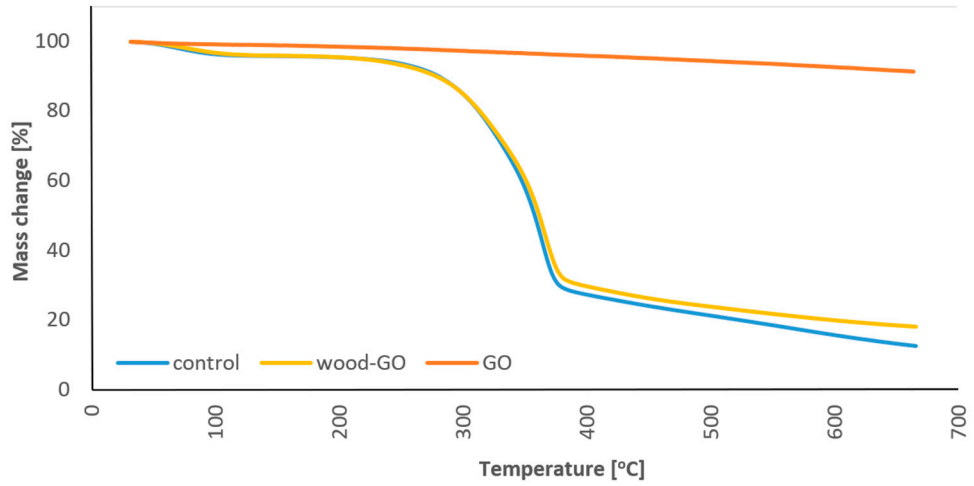
Table 2. Crystallinity of wood modified with graphene oxide.

Samples	Degree of Crystallinity X_c/%
control	57
wood-GO	64

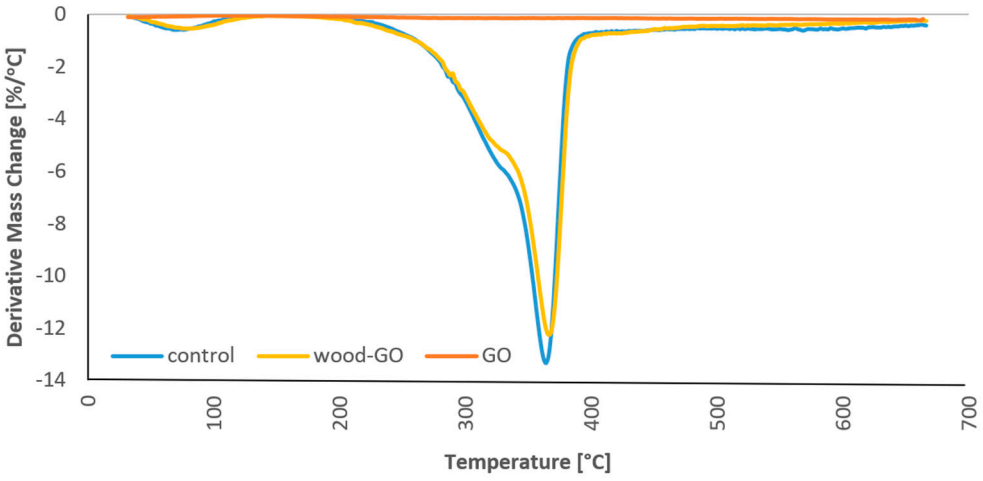
3.1.2. Thermal Properties

The TG/DTG spectra of the GO-impregnated pinewood and control wood are shown in Figure 3a,b. The initial weight loss of the control samples and the samples modified with graphene oxide observed at 100 °C was similar and amounted to less than 4%. The observed changes are primarily related to the evaporation of physically absorbed water and volatile non-structural components [23]. The second mass loss was observed in a temperature range of 270 to 380 °C. The mass loss in this temperature range reaches 70% and is similar in the test and control samples. An endothermic peak at a temperature of about 340 °C corresponds to the thermal decomposition of cellulose into C, CO, CO_2, and H_2O [24]. The wood modified with graphene oxide shows increased thermal stability in a temperature

range of 360 to 660 °C compared with native wood. The DTG curves (Figure 3b) show the peak temperature of maximum thermal degradation, which is approximately 264 °C. Given these DTG thermograms, some improvement in the thermal stability of the GO-modified wood is also visible. According to Shao et al. [25], there are several factors related to the influence of GO on the thermal stability of materials—the occurrence of chemical interactions between GO and cellulose microfibrils and the creation of a kind of structural barrier that prevents or delays the diffusion of decomposition products. The modified wood has higher thermal resistance; however, compared with the control wood, it cannot be said that the practical significance of the modification, from the point of view of fire protection, was achieved with the saturation method used or with respect to the achieved retention of GO in the wood.



(**a**)



(**b**)

Figure 3. Thermogravimetric curves (**a**) and their derivatives (**b**).

3.1.3. Analysis of Microscopic Results

The results of SEM and confocal microscopy indicate the locations of graphene oxide in the wood. Graphene oxide flakes occupy the coil lumens in early and latewood (Figure 4a–d). Significant amounts of GO fill the interior of the resin tubes (Figures 4b and 5a). The strong fluorescence of wood rays in the cross-section indicates the interaction between GO and wood ray cells (Figure 5b). Red arrows in the figures indicate the locations of graphene oxide flakes. Our SEM microscopy studies indicate that graphene oxide does not fill the lumens of all the conductive cells equally. The dense deposition of GO sheets was observed in some

coils and in the lumens of resin conduits. In other places in the cells, the adhesion of single sheets to the surface of the cell wall in the S3 layer was visible (Figure 4a). Based on results obtained by other authors, it can be assumed that there is an interaction between GO and cellulose, which is a component of the wood cell wall [26]. Graphene oxide can achieve high adhesion thanks to conformal contact with the materials with which it is in contact [27]. In turn, Jia et al. [28], based on an assessment of the morphological structure of cellulose fibers, showed that GO adheres to their surfaces, which the authors additionally confirmed with FTIR tests.

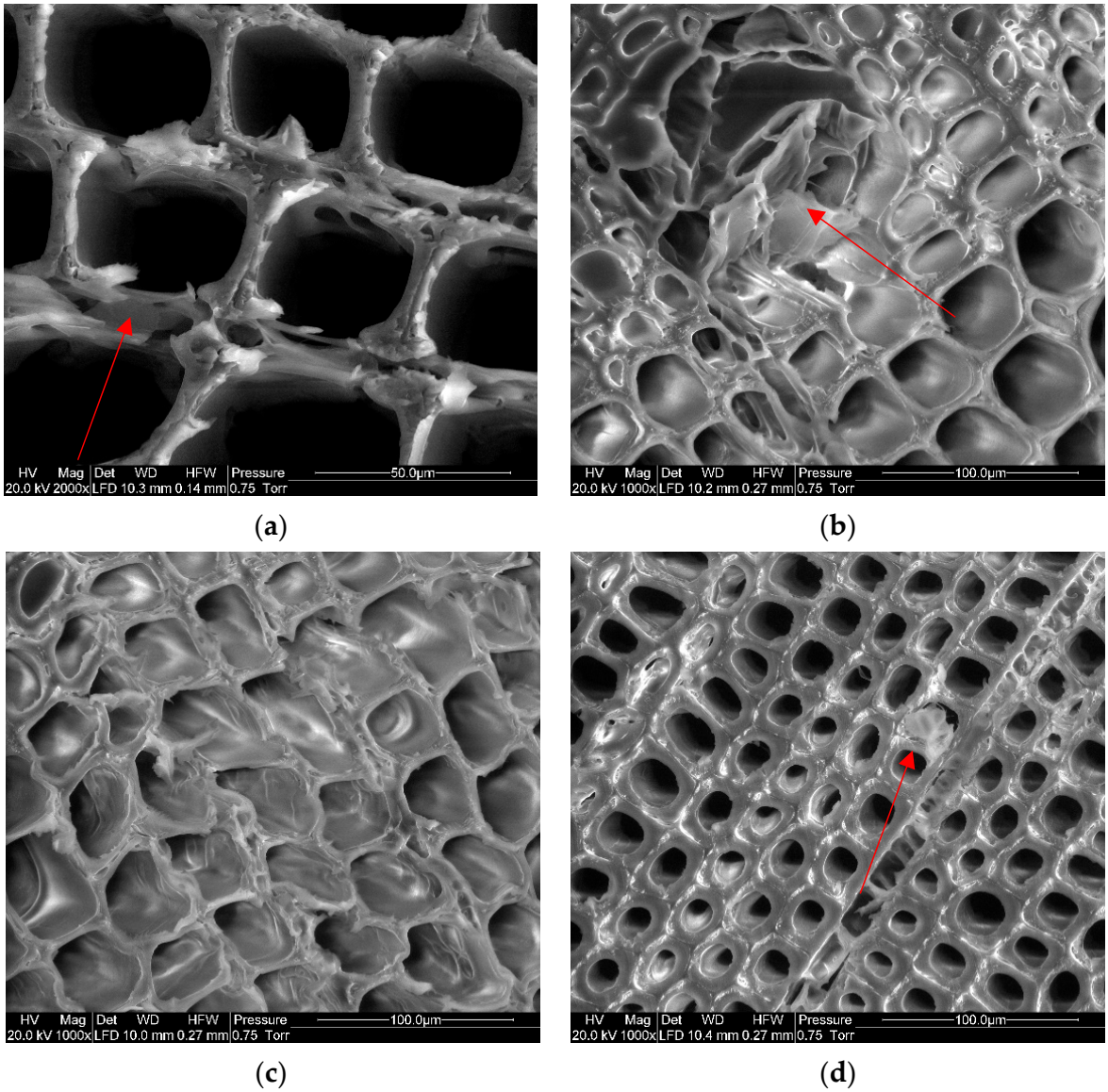
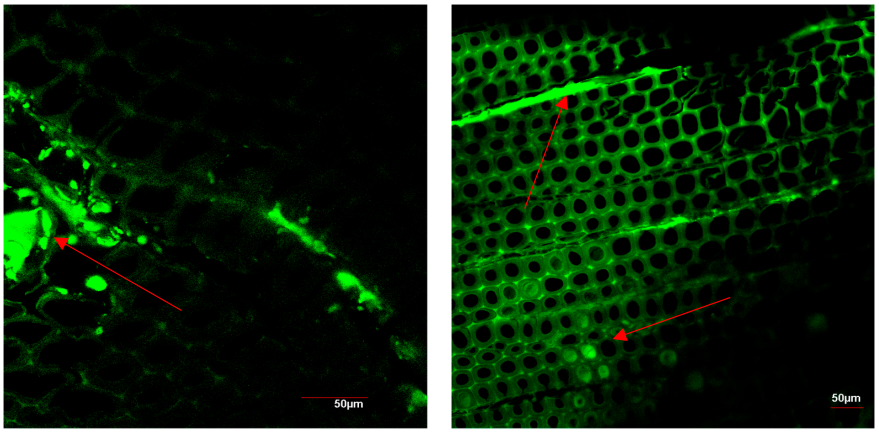


Figure 4. Cross-section of wood—visible layers of graphene oxide flakes in wood cells (**a**–**d**).

Microscopic observations indicate that graphene oxide tightly fills the interior of the anatomical elements of wood, which is visible in SEM images of wood cross-sections (Figure 4b,d). Graphene oxide flakes, despite their small diameter, occupy the entire area of the resin coils and wires, creating complex structures. The reason for this phenomenon may be multiple and result, firstly, from the structure of GO itself, which occurs in the form of 2–3 layered flakes, as well as from the possibility and tendency to create larger aggregates.



(**a**) (**b**)

Figure 5. Fluorescence of graphene oxide labeled with FITC reagent (**a**,**b**).

3.1.4. Test Results for Water Absorption

Wood absorption is a basic parameter influencing dimensional stability and strength parameters. Water destroys wood and promotes the development of biocorrosion. Based on the results obtained, there were no statistically significant differences in the water absorption of the modified or control wood (Table 3), although the average water absorption values indicate a reduction in the mass change curve of samples impregnated with graphene oxide (Figure 6). We showed that the addition of graphene oxide to wood starch composites reduces the water absorption capacity of the composite, and more importantly, the hydrophobicity of the material increases with the increase in the graphene oxide content. It can, therefore, be assumed that the lack of significant differences in wood absorption between the modified and control samples is not due to the nature of graphene oxide but to its low content in wood cells.

Table 3. Water absorption of wood impregnated with graphene oxide.

Samples	*p*-Value	α
	Tukey's Test	
F statistic	0.1674	0.05
control	a	
wood-GO	a	

a—homogeneous groups in the Tukey test; *p*-value—significance of the F statistic; α—statistical significance level.

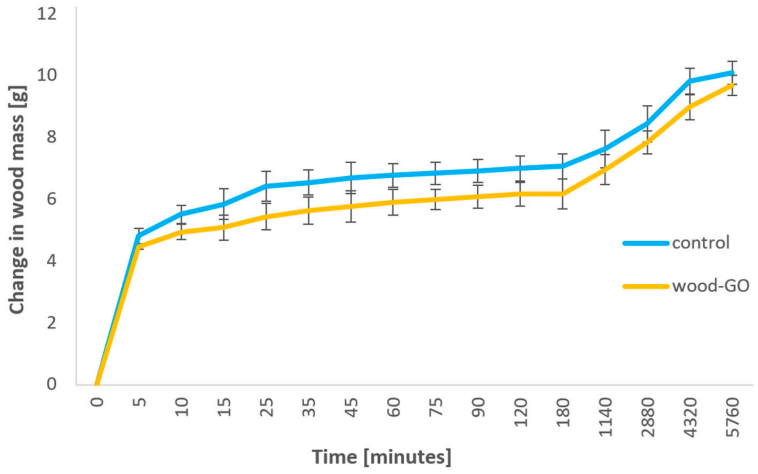


Figure 6. Change in mass of wood samples in contact with water.

3.1.5. Growth of Fungi

The assessment of the degree of mold overgrowth in wood indicates that the GO retention of 0.25 kg/m^3 is not sufficient to protect the wood against molds. Additionally, it was noted that, in a very short time (2 days), the fungus *Trichoderma viride* grew on the entire surfaces of the samples impregnated with graphene oxide. This may suggest that GO is an easy source of carbon, easily assimilable by fungal cells, available on the wood's surface (Figure 7).

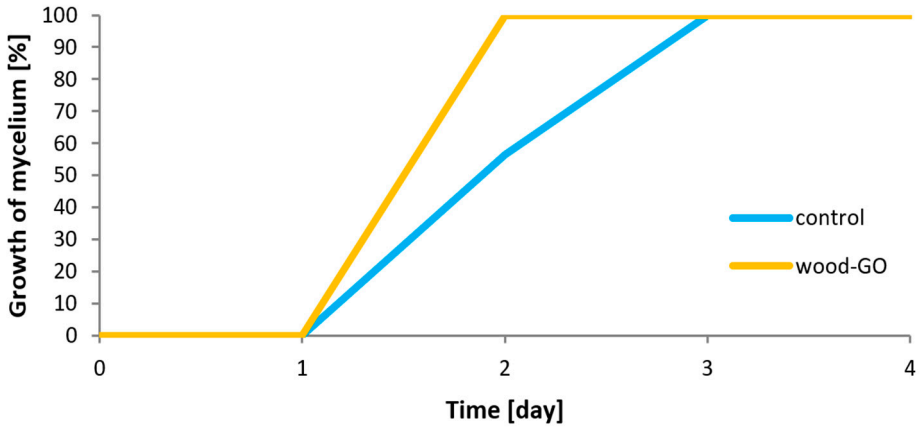


Figure 7. Growth of *T. viride* on a wood surface.

The presence of low concentrations of graphene oxide in the culture medium did not inhibit the growth of the fungus *Trichoderma viride*; however, a decrease in the growth activity of mycelial hyphae was observed with an increasing concentration of nanomaterial in the medium (Table 4). According to research conducted by Nguyen et al. [29], the exposure of fungal cells to graphene oxide causes a reduction in biomass and the abnormal appearance of hyphae. Based on microscopic studies with labeled graphene oxide, it was found that this substance is taken up and metabolized by fungal cells, as evidenced by the fluorescence of their cellular structures (Figure 8a,b). At the same time, it was found that the presence of GO in the culture medium induced changes in the morphological structure. Graphene oxide, as a stress factor, initiates changes at the cellular level characterized by the body's formation of structures called chlamydospores (Figure 8a–d). Blue arrows in the photos indicate the presence of chlamydospores formed at the end of the hyphae. The formation of chlamydospores is the result of the exposure of the fungal hyphae to oxidative stress and a form of defense reaction to difficult growth conditions [30].

Table 4. Growth diameter of *T. viride* on a medium containing various amounts of graphene oxide.

Concentration of Graphene Oxide in Growth Medium (mL/100 mL)	Day of Observation			*p*-Value	α
	2	4	6		
	Growth Diameter of Mycelium (mm)			Tukey's Test	
	F Statistic			8.79×10^{-2}	0.05
0 (control)	69.4	90.0	90.0	a	
0.01	51.0	82.3	83.8	a	
0.05	48.2	77.3	79.3	a	
0.1	48.2	74.7	76.7	b	
0.25	48.2	72.0	74.3	b	
0.5	45.5	70.3	72.7	b	

a, b—homogeneous groups in the Tukey test; *p*-value—significance of the F statistic; α—statistical significance level.

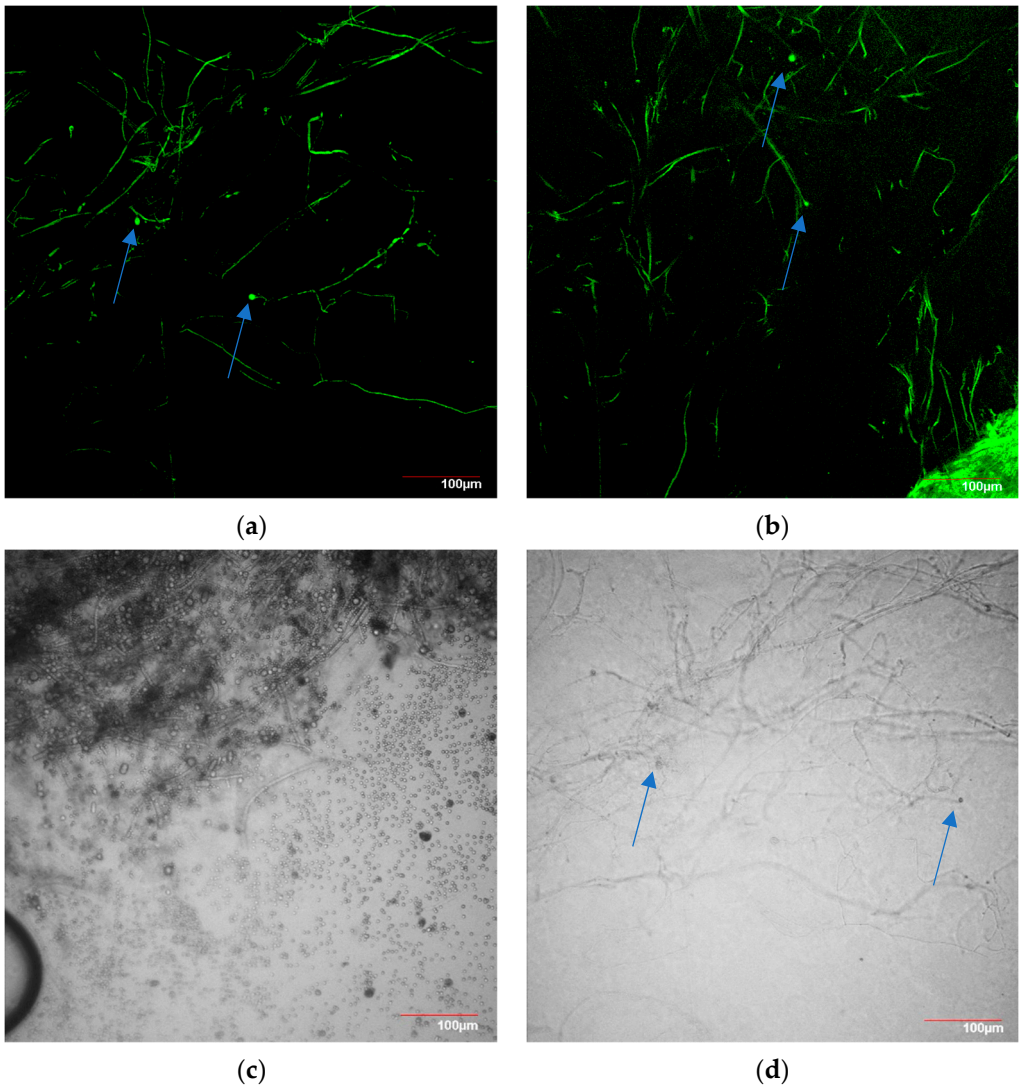


Figure 8. Changes in the morphology of the cellular structures of the fungus *T. viride* under the influence of graphene oxide.

4. Conclusions

The presented work shows that a GO aqueous solution with a concentration of 0.004% is capable of being absorbed by wood in an amount close to the saturation capacity of wood with water, and therefore, no changes in this respect are visible; i.e., the wood can be effectively saturated with GO aqueous solutions. However, an important question arises as to whether the wood will also be effectively saturated at higher concentrations of graphene oxide. The use of such low concentrations of graphene oxide was intended to clarify whether a small amount of graphene oxide effects changes in the properties of wood. With such a low retention rate obtained from saturating the wood in a low vacuum for 30 min, an increase in the crystallinity of the wood and slight changes in its thermal parameters were found. However, no influence of graphene oxide on changes in the hygroscopicity of wood was found. It is, therefore, necessary to conduct further in-depth research on impregnation processes and to determine the retention of graphene oxide in wood, which would result in significant changes in physical and mechanical parameters from the point of use and technology.

Graphene oxide affects the growth and morphology of fungal cells. Low concentrations of GO in the growth medium of the *T. viride* fungus did not completely inhibit growth, but its effect on morphological features was visible in the form of chlamydospores produced by the organism. The appearance of this type of cellular structure is a reaction to

a stress stimulus present in the environment. It can, therefore, be concluded that graphene oxide may have a fungicide function, but to demonstrate this, extended tests should be carried out.

Author Contributions: Conceptualization, I.B.; methodology, I.B., S.J., S.B. and M.K.; software, B.A.; validation, B.A.; formal analysis, I.B.; investigation, P.B.; resources, I.B.; data curation, I.B.; writing—original draft preparation, I.B.; writing—review and editing, B.A., S.B., K.K. and P.B.; visualization, I.B.; supervision, I.B.; project administration, I.B. and P.B.; funding acquisition, P.B. All authors have read and agreed to the published version of the manuscript.

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