Dimitar Angelski*, Krasimira Atanasova

Water Permeability and Adhesion Strength of Bio-based Coating Applied on Wood

Vodopropusnost i adhezivna čvrstoća biopremaza nanesenoga na drvo

ABSTRACT • The main task of wood manufacturers is to ensure the supply of safe wood products. In this respect, the use of plants as raw material for the wood coatings production is a sustainable alternative to fossil fuels, especially since innovative materials minimize the health and environmental risks of the final product over its entire lifespan. This paper presents a study of the water permeability of a water-borne bio-based coating applied to spruce (Picea abies), a study of the adhesion strength of the same coating applied to spruce (Picea abies), beech (Fagus sylvatica L.) and beech plywood, as well as a study of the adhesion strength on spruce after the water permeability test. The tests were performed according to EN 927-5:2006 and EN ISO 4624:2016. The roughness parameters were measured before and after the water permeability test. The coating was found to be hydrophobic but also water permeable. The highest value of adhesion strength was observed for beech surface, the lowest for spruce. The changes in the surface profiles after the water permeability test are insignificant. According to the water absorption criteria, this coating system could only be applied on exterior wood intended for end-use categories such as overlapping cladding, fencing, garden sheds, open cladding, and ventilated rain screen.

KEYWORDS: water permeability; adhesion strength; bio-based coating; beech; spruce


KLJUČNE RIJEČI: vodopropusnost; adhezivna čvrstoća; biopremaz; bukovina; smrekovina

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1 INTRODUCTION

1. UVOD

Wood lignocellulosic biopolymers (cellulose, lignin, hemicelluloses) are subjected to progressive oxidative degradation processes under the action of environmental factors (UV radiation, moisture, heat/cold variations, atmospheric oxygen), which affect wood native durability and cause the occurrence of significant structural and colour changes (discoloration), along with a progressive diminution of its resistance against biological agents (biodegradation or decay development) and its mechanical properties (Teacă et al., 2019). Furthermore, wood is a hygroscopic material that can adsorb or desorb water in response to the temperature and relative humidity of the surrounding atmosphere. Consequently, the moisture content of wood is one of the most important variables affecting its physical and mechanic properties (Hartley, 2001). In this context, wood products are easy to absorb water and steam when they are exposed to environmental conditions for a long time, which greatly affects the durability of wood products and degrades their properties. One effective way to prevent wood degradation processes is to apply coatings on the surface. The varying moisture content of wood results in dimensional and conformational instability, which can compromise the performance of other materials combined with wood, such as adhesives and surface coatings (Man tanis and Papadopoulos, 2010). For protective and decorative reasons, coatings (varnishes) are generally applied on wooden substrates as multi-layered systems that are composed of primer and topcoat. Their application on wood materials aims to enhance the biological, physical, and mechanical properties of wood.

The wood coatings industry represents an active field of research and development, driven by the necessity to produce high-performance materials able to respond to different environmental regulations and constraints. Recently, the attention is turning towards the development of coatings based on green materials, namely “bio-based coatings”, obtained through sustainable processes that do not generate toxic emissions (Sarcinella and Frigione, 2023). Recent trends in this area include the use of bio-based natural products (such as wood and plant extractives, vegetable oils, natural waxes, different biopolymers, and biological control agents) and nano-based materials (Teacă et al., 2019). For all these products, the main criterion for the estimation may be represented by the protection provided by the physical and mechanical properties of coatings applied on the wood surface. The basic properties of a coating are determined by its main components, namely binders, pigments, solvents, fillers, and additives (Sandberg, 2016). Permeability of coatings for water and water vapour is a supreme factor in their wood protective function. Permeability is a measure of the ease with which fluids are transported through a porous solid under the influence of a pressure gradient (Siau, 2012). Liquid water and water vapour uptake are affected by coating film thickness, number of coats, and coating composition.

In order to perform satisfactorily, coatings must adhere to the substrates on which they are applied. Adhesion is a complex physico-chemical phenomenon for which, however, there is not a rigorous theoretical definition. Adhesion is difficult to define, and an entirely satisfactory definition has not yet been found (Silva et al., 2011, Ebnesajjad and Landrock, 2014). Therefore, a quantitative analysis of coating adhesion on wood is needed. One of the main factors used for the evaluation of the adhesion of coatings is the adhesion strength. Adhesion strength is an important feature to measure the durability of the coatings. Various methods are used to evaluate adhesion strength of the coats. The most objective and widespread method is the pull-off test. The main advantage of this method includes its practicality and simple application for different surfaces. Adhesion of a single coating or a multi-coat system of paint, varnish or related products is assessed by measuring the minimum tensile stress necessary to detach or rupture the coating in a direction perpendicular to the substrate.

In furniture production, the coating processes have great importance for technical, economic, aesthetic, and ecological evaluation of the wood materials. Different coating technologies provide different levels of protection. More than 95 % of exterior wood coatings are applied as liquid coatings, either solvent- or water-borne (e.g. acrylic, polyurethane, alkyd), but their use and subsequent emission of volatile organic compounds (VOCs) represents a dominant concern because of their significant contribution to global warming (Teacă et al., 2019). As the coatings industry seeks to improve its environmental impact, plant-based technologies present a major innovation opportunity. Natural extractives, vegetable and essential oils, natural resins and waxes, and various biopolymers can be included in coating formulations (primarily water-borne ones) to prevent or limit the harmful impact of these formulations on the environment due to their specific properties (e. g. non-toxicity, reversible character, resistance to moisture and solvents, compatibility). The aim of this study is to determine the water permeability and adhesion strength of bio-based coating for the protection of wood surfaces. Therefore, the axial pull-off test was used to determine the adhesion strength of samples manufactured from different wood species to provide an initial data for improving the quality of finished products manufactured from bio-based raw mate-
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2 MATERIALS AND METHODS

For the tests, spruce (Picea abies) with an average density of 440 kg/m³, European beech (Fagus sylvatica L.) with an average density of 730 kg/m³ and beech plywood with an average density of 750 kg/m³ were used. The plywood surface was sanded with a P80-120 grain size sandpaper supplied by the manufacturer (S. C. Cildro Plywood, Romania).

Two sets of test specimens were prepared. The samples from spruce with dimensions 150 mm × 70 mm × 20 mm (longitudinal, tangential, and radial directions, respectively) were prepared for the water permeability test. The test samples were manufactured in accordance with the specifications in EN 927-5:2006. The specimens were chosen for their lack of defects, such as knots, cracks, or resin spots.

For the adhesion strength test, samples of spruce and beech wood with dimensions 150 mm × 50 mm × 20 mm and beech plywood with dimensions 50 mm × 150 mm × 15 mm were prepared. All samples were conditioned prior to coating application (temperature of (20±2) °C and relative humidity of (65±5) %).

For coating of samples, a bio-based wood stain with the colour “tobacco” for outdoor application, produced by Industrias Quimicas Masquelack, S. A. (Spain), was used (Figure 1). According to the information given by the manufacturer, it creates a water-resistant, breathable and flexible film with high outdoor durability. The coating system is water-borne with a bio-based content of 39 % and contains Discovery® SP - 7450 by Covestro. Discovery® SP - 7450 is an acrylic copolymer emulsion with elongation, blocking resistance, and early water resistance that is suitable for clear and opaque formulations in high-demand outdoor durability applications. The bio-based wood stain mixtures had a viscosity of 21 s when measured at 23 °C in a DIN 4 mm viscosity cup.

The test samples were sanded with P150 grain size sandpaper, and the first layer of the bio-based wood stain was applied by brush. After drying, the surfaces were treated with abrasive steel wool (Scotch-brite®) and a second layer was applied. The stain amounts $Q$ for the two layers were $Q_1 = 80$ g/m² and $Q_2 = 30$ g/m², respectively. After the coating drying, the average dry residue content of hardened film is 17.34 %.

To establish the coating hydrophobicity, the behaviour of drops of distilled water on the surfaces of the various test specimens was observed. The drops are dosed with a micropipette and have a volume of 2.5 µm.

The European standard EN 927-5:2006 specifies a test method for assessing the liquid water permeability of coating systems for exterior wood by measuring the water absorption of coated wood panels. After the application of the coating, all the remaining sides were sealed with a two-component polyurethane (PU) system. The PU system has a predetermined water permeability of 16 g/m². For each set of six replicates, the arithmetic mean value of the weight increase and the standard deviation were calculated. The arithmetic mean value of the weight increase after 72 hours of floating is reported as the mass of absorbed water per test face area. The amount of water absorbed by each sample was measured as mass of absorbed water ($MWA$) per test face area relative to the weight of the conditioned specimen prior to the test, Eq. (1):

\[
MWA_i = \frac{(w_i - w_o)}{A}
\]

Where $MWA_i$ – mass of absorbed water per area in g/m² at time 1; $w_o$ – weight in g at time 0; $w_i$ – weight in g at time 1; $A$ – area of test face in m².

The adhesion strength of the coating is defined by a standardized pull-off method (ISO 4624:2016) with a glued stamp (dolly). The test is performed by securing a loading fixture (dolly) perpendicular to the surface of the coating with an adhesive. After the adhesive is cured, a testing apparatus is attached to the loading fixture and aligned to apply tension perpendicular to the test surface. The force applied is gradually increased and monitored until either a plug of coating material is detached, or a specified value is reached.

Figure 1 Wood surfaces coated with bio-based wood stain: A – spruce (Picea abies); B – European beech (Fagus sylvatica L.); C – beech plywood

Slika 1. Drvene površine premazane biolazurom: A – smrekovina (Picea abies); B – bukovina (Fagus sylvatica L.); C – bukova furnirska ploča
The Mitutoyo SJ-210 surface roughness measurer (Mitutoyo, Japan) was used to indicate the surface roughness at the following settings: profile – $R$ (radius - $5 \mu m$), filter - Gauss; number of segments $N = 6$; cut-off length $\lambda c = 2.5 \, mm$; measuring speed – $0.25 \, mm/s$. The measurements were made perpendicular to the wood grains. Surface parameters like arithmetic mean deviation of the assessed profile ($Ra$), maximum height of profile ($Rz$), maximum profile peak height ($Rp$), total height of profile ($Rt$), maximum profile valley depth ($Rv$) and mean width of the profile elements ($RSm$) were estimated according to ISO 4287:1997. Roughness depth ($Rk$) reduced peak height ($Rpk$), as well as reduced valley depths ($Rvk$) were estimated by ISO 13565-2:1996.

3 RESULTS AND DISCUSSION
3. REZULTATI I RASPRAVA

3.1 Water permeability of bio-based coating
3.1. Vodopropusnost biopremaza

The obtained bio-based coating is hydrophobic because the static water contact angle $\theta$ is $> 90^\circ$ (Figure 2). The water permeability test, on the other hand, revealed that the bio-based coating applied on the spruce specimens showed extremely high water-permeability (the average value of $725 \, g/m^2$).

The time-dependent mass of absorbed water ($MWA$) during the water permeability test of spruce ($Picea abies$) coated with bio-based stain is shown in Figure 3. Manufacturers mention that the used bio-based wood stain is water resistant, but the two coats applied were completely insufficient. According to the water absorption criteria in EN 927-2:2022, this coating system is non-stable. Mostly, this is due to the porous, breathable, and thin protective coating made by bio-based stain.

It is also possible to free the movement of coated wooden products. By classification in EN 927-1:2013, this system could be used outdoor only for end-use categories such as overlapping cladding, fencing, garden sheds, open cladding, and ventilated rain screen.

After the water permeability test, the surface profiles of the investigated samples have changed. A quantitative assessment of these changes is given by the roughness parameters. The changes in the roughness parameters in the selected study phases indicate the redistribution mechanism of the remaining water in the test specimen.

Table 1 shows the average roughness parameters of a surface coated with bio-based stain before and after water permeability testing. The table also presents the percentage changes in the roughness parameters after 72 hours of drying the samples at room temperature ($20 \pm 2 \, ^\circ C$) and relative humidity ($65 \pm 5 \%$). Measurements of roughness parameters were made on the
same sections of the same length. The values show a decrease in the roughness of the coating after the water permeability test, with the largest percentage decrease in the parameters \( R_p \) (maximum profile peak height) and \( R_{pk} \) (reduced peak height). This means that the coating is elastic enough to act as a barrier which limits grain raising.

Based on the high water-permeability of the studied coating, it can be concluded that using the bio-based stain by itself will not provide proper water protection of the wood products. It is advisable to apply an acrylic topcoat with proven hydrophobic properties.

### 3.2 Adhesion strength of bio-based coating

**3.2. Adhezivna čvrstoća biolazure**

After the pull-off test according to ISO 4624:2016, (90-100) % adhesion destruction was observed between the substrate and the coating (Figure 4). According to the wood species (Figure 5), the adhesion strength of stain is higher on European beech (Fagus sylvatica L.) and on beech plywood than on spruce (Picea abies). These results confirm the general statement that the adhesion strength of coatings is higher on hardwood than on softwood (Kaygın and Akgun, 2008). There are a lot of factors that may cause this difference among the species, e.g. intensity, cell structure, basic and secondary compounds of wood, texture, extractive substances (Kaygın and Akgun, 2008).

The two-layer bio-based coating on wood surfaces has sufficiently good adhesion strength (Figure 5). According to the technological criteria for outdoor paints and varnishes (Decision, 2009), the critical value of the adhesion strength is 1.5 N/mm².

Adhesion of coatings on wood is most critical under wet conditions (de Meijer, 1999; Sönmez et al., 2009). In this regard, the adhesion strength of the spruce specimens was measured after the water permeability test, i.e. after 72 h of sorption and 144 h of desorption. The average value obtained for the adhesion strength of the spruce specimens after the water permeability test is 2.58 N/mm², which is a decrease of 2.7 %. This means that the coating of bio-based stain

### Table 1

<table>
<thead>
<tr>
<th>Roughness parameter</th>
<th>Average values, µm</th>
<th>Accuracy index, %</th>
<th>Change, %</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Srednje vrijednosti, µm</td>
<td>Indeks preciznosti, %</td>
<td>Promjena, %</td>
</tr>
<tr>
<td>Ra</td>
<td>4.84 4.77 4.87</td>
<td>2.16 2.22 2.11</td>
<td>-1.57 2.22</td>
</tr>
<tr>
<td>Rz</td>
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<td>1.80 1.87 2.09</td>
<td>-4.13 -0.03</td>
</tr>
<tr>
<td>Rp</td>
<td>16.81 15.51 15.48</td>
<td>1.90 2.06 2.23</td>
<td>-7.76 -0.17</td>
</tr>
<tr>
<td>Rv</td>
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<td>2.11 2.10 2.37</td>
<td>-0.91 0.09</td>
</tr>
<tr>
<td>Rsm</td>
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<td>-4.58</td>
</tr>
<tr>
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</tr>
<tr>
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<td>2.50 2.49 2.32</td>
<td>-1.52 4.63</td>
</tr>
<tr>
<td>Rpk</td>
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<td>2.39 3.23 3.35</td>
<td>-9.84 1.29</td>
</tr>
<tr>
<td>Rvk</td>
<td>7.52 7.67 7.35</td>
<td>2.31 2.72 3.00</td>
<td>1.95 -4.19</td>
</tr>
</tbody>
</table>

**Figure 4** Typical destruction after axial pull-off test

**Slika 4.** Tipični lom nakon pull-off testa

**Figure 5** Adhesion strength of wood samples coated with two-layer bio-based stain

**Slika 5.** Adhezivna čvrstoća uzoraka drva premazanih dvama slojevima biolazure
Applied to wood retains almost all its adhesion strength after contact with water.

4 CONCLUSIONS
4. ZAKLJUCAK

Under the conditions of this study and based on the results obtained, the following conclusions can be made specifically for the above-mentioned materials:

Good hydrophobicity of the studied bio-based stain applied to wood has been established;

The water permeability of the studied two-layer bio-based coating is 725 g/m². According to the water absorption criteria, this coating system is non-stable. This coating system could be used outdoor only for end-use categories such as overlapping cladding, fencing, garden sheds, open cladding, and ventilated rain screen;

The two-layer bio-based coating on wood surfaces has comparatively high adhesion strength and retains almost all of it after 72 hours of contact with water.

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5 REFERENCES
5. LITERATURA


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