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Development of Laminated Flooring Using Wood and Waste Tire Rubber Composites: A Study on Physical-Mechanical Properties

Izrada laminata upotrebom kompozita od drva i otpadne gume: studija o fizičko-mehaničkim svojstvima

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ABSTRACT • This study aimed to develop laminate flooring composite using a combination of wood and waste tire rubber (WTR). Plywood panels were produced by using beech (*Fagus orientalis*), alder (*Alnus glutinosa*), and poplar (*Populus*) veneers in a 7-ply configuration. To enhance the physical-mechanical properties of the panels, three loadings of nano-SiO₂ (0, 2, and 4 wt%) along with 2 wt% of hexamethyldisilazane (HMDS) were added. Commercial urea-formaldehyde (UF) resin and methylene diphenyl diisocyanate (MDI) were used to bind the wood layers and rubber layers together. The mechanical properties, including modulus of rupture (MOR), modulus of elasticity (MOE), impact strength (IS), hardness strength (HS), and physical properties, such as density (D), water uptake (WU), and thickness swelling (TS), were evaluated. The results showed that increasing the WTR content led to improvements in the physical properties (D, WU, and TS), while negatively affecting the mechanical properties (MOR, MOE, IS, and HS) of the resulting panels. However, the addition of nano-SiO₂ improved both the physical and mechanical properties (MOR, MOE, and HS) of the panels. Furthermore, it was observed that the mechanical properties were enhanced with increasing the number of beech layers, although the WU of panels decreased compared to panels made with alder and poplar. Overall, the improvement in the physical properties of the panels followed the order of the arrangement of rubber layers > nano-SiO₂ content > veneer layers.

KEYWORDS: silica nanoparticles; rubber waste; plywood; physical-mechanical properties; flooring

SAŽETAK • Postavljeni je cilj bio izraditi kompozitni laminat kombinacijom drva i otpadne gume (WTR). Furnirske su ploče proizvedene upotrebom sedam slojeva furnira drva bukve (*Fagus orientalis*), johe (*Alnus glutinosa*)

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i topole (Populus). Kako bi se poboljšala fizička i mehanička svojstva ploča, dodana su tri punjenja nano-SiO₂ (0, 2 i 4 wt%), zajedno s 2 wt% heksametildisilazana (HMDS). Za lijepljenje slojeva drva i gume rabljena su komercijalna urea-formaldehidna (UF) smola i metilen difenil diizocijanat (MDI). Zatim su proučavana mehanička svojstva tako dobivenih laminata uključujući modul loma (MOR), modul elastičnosti (MOE), udarnu čvrstoću (IS), otpornost na zasijecanje (HS) i fizička svojstva kao što su gustoća (D), upijanje vode (WU) i debljinsko bubrenje (TS). Rezultati su pokazali da je povećanje sadržaja WTR-a pridonijelo poboljšanju fizičkih svojstava dobivenih ploča (D, WU i TS), dok je negativno utjecalo na njihova mehanička svojstva (MOR, MOE, IS i HS). Međutim, dodatkom nano-SiO₂ poboljšana su fizička i mehanička svojstva ploča (MOR, MOE i HS). Nadalje, uočeno je da su mehanička svojstva poboljšana povećanjem broja slojeva bukovine, iako su ploče od bukovine upijale manje vode u usporedbi s pločama od drva johe i topole. Općenito, poboljšanje fizičkih svojstava ploča slijedilo je načelo: raspored slojeva gume > sadržaj nano-SiO₂ > slojevi furnira.

KLJUČNE RIJEČI: nanočestice silicija; gumeni otpad; furnirska ploča; fizikalno-mehanička svojstva; podovi

1 INTRODUCTION

1. UVOD

The world has agreed to move from a linear to a circular economy. This has led to various inventions, new business models, and strategies. They support the push for further institutionalization of circular economy practices. Following globalization, more waste is produced, surpassing the earth's capacity for renewal and natural resilience (Greer *et al.*, 2021). For example, it is estimated that the annual production of waste in the world will reach 50-70 % by 2050 (Kaza *et al.*, 2018). Thus, it is of great interest to considerably reduce the amounts of produced waste and to develop alternative strategies to manage wide spectra of wastes considering the attitudes of the circular economy. One of the materials that offer a lot of opportunities for recycling and reuse is rubber. Currently, rubbers are mainly being used to fabricate a wide range of materials for numerous applications like toys, automotive, technical, construction and structures, etc. However, rubber does not degrade at the end of its life and can remain in the environment for decades, predominately due to the presence of reinforcing fillers, antioxidants, antiozonants, and curating agents (Fazli and Rodrigue, 2020). The landfilling and combustion are not in accordance with the circular economy principles or with the waste management hierarchy (Pires and Martinho, 2019). Therefore, it is potentially valuable to develop alternative strategies for the reuse and recycling of rubber. One of the possible alternatives is wood-rubber composites (Shao *et al.*, 2016). The idea of these composites is to combine wood with superior mechanical properties and a good density to strength ratio (Madsen and Gamstedt, 2013) with rubber. On the other hand, the benefits of the rubber are high compressive performance, good water sorption properties, absorption of damping vibration reduction, unique energy absorption, good toughness, strong abrasion resistance, low biodegradability, etc (Mancel *et al.*, 2022). Therefore, producing composites based on wood and rubber could have superior properties (Ayrilmis *et al.*, 2009). There are various approaches how to pro-

duce wood-rubber composites. Wood can be used as a filler in the rubber (Nuzaimah *et al.*, 2018; Flour *et al.*, 2004; Vladkova *et al.*, 2004; Vladkova *et al.*, 2006), and rubber can be used in particleboards (Mancel *et al.*, 2022) or oriented strand boards (Ayrilmis *et al.*, 2009a). The last but not least alternative is to use rubber as a layer between the veneers in plywood. In such applications, rubber can even serve as an adhesive (Shao *et al.*, 2016). Ashori *et al.* (2015) investigated the possibility of using waste tire rubber in fabricating hybrid plywood composite panels. The results indicate that utilizing rubber in the produced panels enhanced physical properties such as water uptake (WU), thickness swelling (TS), and sound absorption. However, mechanical properties like modulus of rupture (MOR), modulus of elasticity (MOE), and impact strength (IS) were declined. Recently, nanotechnology has improved the efficiency of materials in different areas of science. Among nanoparticles, nano-silica (SiO₂) with very small sizes and high surface area are widely used in the polymer and surface coating industries (Fang *et al.*, 2014; Xiong *et al.*, 2021). Plenty of studies have focused on the application of nanosilica in numerous polymers. However, only a few studies refer to wood/rubber and wood/plastic composites. It has been reported that nano-SiO₂ can improve polymer matrix mechanical properties such as strength, hardness, modulus of crystallinity as well as viscosity and adhesion (Parvinzadeh *et al.*, 2010). Deka and Maji (2013) investigated the influence of nanoclay and nano-SiO₂ on the properties of wood/plastic composites. The results showed that mechanical properties, thermal properties, and flame retarding were improved by increasing nanoparticles by up to 3 wt% of each nanoparticle. The impact strength of wood /plastic composite decreased with the increase of clay and nano-SiO₂ (3 and 4 wt%) (Nourbakhsh *et al.*, 2011). Xu and Zhang (2021) investigated the preparation and properties of hydrophobically modified nano-SiO₂ nanoparticles with hexadecyltrimethoxysilane (HDTMS). Nano-SiO₂ is a common inorganic silicon material. However, its surface is very hydrophilic due to the presence of many hydroxyl groups, which limits its use in some

fields. Therefore, in their research, they used HDTMS to modify nano-SiO₂. The results of water contact angle analysis (WCA), Fourier transform infrared (FTIR), two-dimensional correlation spectroscopy (2D-COS), thermal analysis (TG), and scanning electron microscopy (SEM) showed that HDTMS was successfully grafted on the nano-SiO₂ surface due to its long hydrophobic alkyl chain. In general, their results showed that, when the ratio of nano-SiO₂ and HDTMS was 0.25:1, the WCA reached 170.9°, which was about 5.62 times higher than before the modification. In fact, it showed the property of superhydrophobicity (Xu and Zhang, 2021).

This research attempted to develop a more sustainable and innovative rubber-based plywood by optimizing its composition, employing structural engineering principles, and incorporating additional nano-SiO₂. By utilizing advanced technology and innovative materials, this research aims to contribute to the growing field of sustainable building materials and promote the adoption of environmentally friendly alternatives in the construction industry.

2 MATERIALS AND METHODS

2. MATERIJALI I METODE

2.1 Materials

2.1.1. Materijali

Rotary-cut veneers of beech (*Fagus orientalis*), alder (*Alnus glutinosa*) and poplar (*Populus alba* L.) with densities of 0.68 g/cm³, 0.49 g/cm³ and 0.40 g/cm³, respectively, were used for the work. The mentioned wood species are the most dominant species used for plywood manufacturing in Iran. A total of 288 defect-free veneer samples with a dimension of 400 mm × 400 mm and a thickness of 1.8 mm were condi-

tioned to a moisture content of 10 % prior to experiments. The chemical materials used, their characteristics and suppliers are listed in Table 1 for easy reference.

2.2 Methods

2.2.1. Metode

2.2.1 Preparation of nanoparticle soluble polymer

2.2.1.1. Priprema polimera s otopljenim nanočesticama

A combined solution of nano-SiO₂ and HMDS was used to reinforce the rubber powder filler (Figure 1). In fact, distilled water and ethanol were used as solvents in the reaction, crosslinking and providing proper stabilization of the silica particles. To prepare the nanoparticle solution with suitable concentrations, nano-SiO₂ powder with weight ratios of 0, 2, and 4 wt% and HMDS with 2 wt% dry weight ratio of rubber powder were added to 150 L of a mixture of distilled water and ethanol and placed in a magnetic electric mixer and combined for 2 h. The combined solution of nano-SiO₂ and HMDS was then placed in a drying chamber (laboratory dryer) at 8 °C for 8 h to obtain a polymer nanoparticle solution. Finally, the generated residues were mixed as a dry powder tablet and mixed with rubber powder (Song *et al.*, 2014).

2.2.2 Hybrid plywood composite fabrication

2.2.2.1. Izrada kompozita od hibridne furnirske ploče

Table 2 summarizes the features of some seven-layer plywood samples, comprising a combination of beech (B), alder (A), and poplar (P) veneers with rubber particles. Plywood panels were made using a wide range of permutations across four independent variables: layering pattern, WTR content, nano-SiO₂ content, and hot-

Table 1 Chemical materials used in the study

Tablica 1. Kemikalije upotrijebljene u istraživanju

| Chemical material <i>Kemikalija</i> | Characteristics <i>Svojstva</i> | Supplier (Country) <i>Dobavljač (država)</i> |
|--|--|---|
| Waste tire powder (WTP) <i>prah od otpadnih guma</i> | Bulk density / <i>nasipna gustoća</i> : 0.32 g/cm ³ Particle size / <i>veličina čestica</i> : 0.2 mm | Isatis Yazd (Iran) |
| nano-SiO ₂ | Bulk density / <i>nasipna gustoća</i> : <0.10 g/cm ³ True density / <i>stvarna gustoća</i> : 2.4 g/cm ³ Particle size / <i>veličina čestica</i> : 20-30 nm Purity / <i>čistoća</i> : > 99 % | Evonik Industry (Germany) |
| Hexamethyldisilazane (HMDS) <i>heksametildisilazan (HMDS)</i> | Bulk density / <i>nasipna gustoća</i> : 0.77 g/cm ³ Purity / <i>čistoća</i> : > 97 % Boiling point / <i>vrelište</i> : 126 °C | Tokyo Chemical Industry (Japan) |
| Ethanol / <i>etanol</i> | Bulk density: 0.79 g/cm ³ Purity / <i>čistoća</i> : > 99 % Boiling point / <i>vrelište</i> : 78.5 °C | Scharlau Chemical Industry (Spain) |
| Methylene diphenyl diisocyanate (MDI) resin <i>metilen difenil diizocijanat (MDI)</i> | True density / <i>stvarna gustoća</i> : 1.27 g/cm ³ Solid content / <i>sadržaj suhe tvari</i> : 100 % | Aras Chemicals (Iran) |
| Urea formaldehyde (UF) resin <i>urea-formaldehidna (UF) smola</i> | True density / <i>stvarna gustoća</i> : 1.26 g/cm ³ Solid content <i>sadržaj suhe tvari</i> : 60 % | Iran-Choob Ghazvin (Iran) |

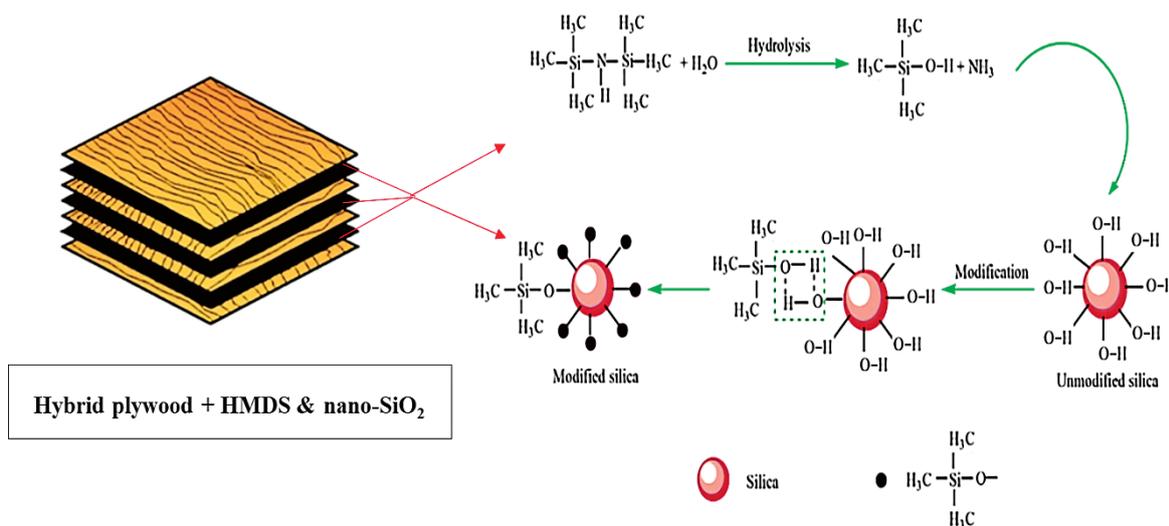


Figure 1 Schematic diagram showing influence of nano-SiO₂ modified by HMDS on bonding of rubber between wood layers (Song *et al.*, 2014)

Slika 1. Shematski dijagram utjecaja nano-SiO₂ čestica modificiranih HMDS-om na lijepljenje gume između slojeva drva (Song *et al.*, 2014.)

pressing method (Table 2). The veneers were cold-pressed for 10 minutes after being bonded with UF resin at a glue spread rate of 160 g/m². It should be noted that MDI glue spread rate was 150 g/m². The plywood was produced using a two-stage hot-pressing technique under controlled conditions. To begin, a molding frame (400 mm × 400 mm) was filled uniformly with resin rubber powder. The particles served as the panel core or intermediate layer after being subjected to high temperatures during the pressing process. The nominal thickness of these rubber layers was 3 and 5 mm and the density 0.9 g/cm³. As a result, the rubber layer and wood veneers were pressed for 10 min at 145 °C and 5 MPa pressure. In order to prepare the hybrid plywood panels for testing, they were conditioned for 7-10 days in a conditioning room with a relative humidity of 65 % and a temperature of 25 °C. Conditioned panels have a more consistent moisture levels and fewer drying strains.

Three identical plywood panels were made for each group. In addition, each veneer layer was 1.8 mm thick, the thickness of the core layer of the rubber was 5 mm and its two sides were 3 mm.

2.3 Physical properties

2.3.1 Fizička svojstva

Density (*D*), water uptake (*WU*), and thickness swelling (*TS*) of each specimen were measured as physical parameters according to EN 323 standard and EN 317 standard, respectively. To condition the composites, the samples were submerged in distilled water at room temperature for 2 and 24 h. After drying, their weight and thickness were re-measured. The samples were immediately measured to the nearest 0.001 mm and weighed to the nearest 0.01 g. All derived values were obtained as the median of six independent calculations.

Table 2 Design of plywood sets

Tablica 2. Dizajn uzoraka furnirske ploče

| Type <i>Vrsta</i> | Sheet set <i>Sklop listova</i> | Layers model <i>Raspored slojeva</i> | Description / Opis | Total thickness, mm <i>Ukupna debljina, mm</i> | Construction <i>Konstrukcija</i> |
|---|-----------------------------------|---|--|---|-------------------------------------|
| Hybrid plywood (PWH) <i>hibridna furnirska ploča</i> | 4-Ply/3-R | VA/R/V/R/V/R/VA VB/R/V/R/V/R/VB VP/R/V/R/V/R/VP | Veneer (V)/Rubber Layer (R)/ Veneer (V)/Rubber Layer (R)/ Veneer (V)/Rubber Layer (R)/ Veneer (V) | 18.2 | V-R-V-R-V-R-V |
| Core plywood (PWC) <i>furnirska ploča s jezgrom</i> | 6-Ply/1-R | 3VA/R/3VA 3VB/R/3VB 3VP/R/3VP | 3 Veneer (V)/1 Rubber Layer (R)/3 Veneer (V) | 15.8 | V ₃ -R-V ₃ |
| Plywood veneer (PWV) <i>furnirska ploča</i> | 7-Ply | REA REB REP | Reference Plywood (Beech-Alder-Poplar) | 12.6 | V ₇ |

Table 3 Properties, dimensions, and number of composite hybrid plywood samples
Tablica 3. Svojstva, dimenzije i broj uzoraka kompozitne hibridne furnirske ploče

| Property test <i>Ispitivano svojstvo</i> | Dimensions, mm <i>Dimenzije, mm</i> | Number <i>Broj uzoraka</i> | Reference standard <i>Referentni standard</i> |
|---|--|-------------------------------|--|
| Density (D) / <i>gustoća (D)</i> | 50×50 | 6 | EN 323:1993 |
| Water uptake (WU) and thickness swelling (TS) <i>upijanje vode (WU) i debljinsko bubrenje (TS)</i> | 50×50 | 6 | EN 317:1993 |
| Flexural strength (FS) and flexural modulus (FM) <i>čvrstoća na savijanje (FS) i modul elastičnosti pri savijanju (FM)</i> | 282×50 346×50 | 6 | ASTM-D 790:2002 |
| Impact bending strength (IBS) / <i>čvrstoća na udar (IBS)</i> | 280×20 | 6 | DIN 52189:1981 |
| Hardness strength (HS) / <i>otpornost na zasijecanje (HS)</i> | 50×50 | 6 | EN 1534:2010 |

2.4 Mechanical properties

2.4. Mehanička svojstva

The samples were conditioned at 20 °C and 65 % relative humidity, and then their modulus of rupture (MOR) and modulus of elasticity (MOE) were measured using ASTM-D 790 standard. At a span-to-depth ratio of 20:1, flexural tests were performed using the third point loading method. Universal testing equipment (Zwick/Roell Z150, Germany) was used to apply a load to the specimen at a strain rate of 4 mm/min. The hardness strength (HS) and the impact bending strength (IBS) of the uncut composite were measured using a pendulum impact tester. Six replications were done for each type of plywood panel. Table 3 lists the dimensions and number of composite hybrid plywood samples used for physical and mechanical tests.

2.5 Statistical analysis

2.5. Statistička analiza

In a completely random setup, $n=3$ samples were analyzed using the statistical software SPSS to see

what happened (version 20.0). The analysis of variance (ANOVA) and Duncan's Multi-Range test (at a 99 % confidence level) were used to compare the means and standard deviations of the groups.

3 RESULTS AND DISCUSSION

3. REZULTATI I RASPRAVA

3.1 Physical properties

3.1. Fizička svojstva

3.1.1 Density (D)

3.1.1.1. Gustoća (D)

According to the ANOVA results, the density of the plywood/rubber samples increased significantly with the addition of 4 wt% nano-SiO₂ (99 % confidence level; Table 1). Duncan's test showed that this treatment had the highest density among all groups, while the plywood samples had the lowest density (Supplement Table 2 and 3). The pressure in the hot press and wood species considerably impacts the density of plywood. The highest density was observed for beech plywood (0.61 g/cm³),

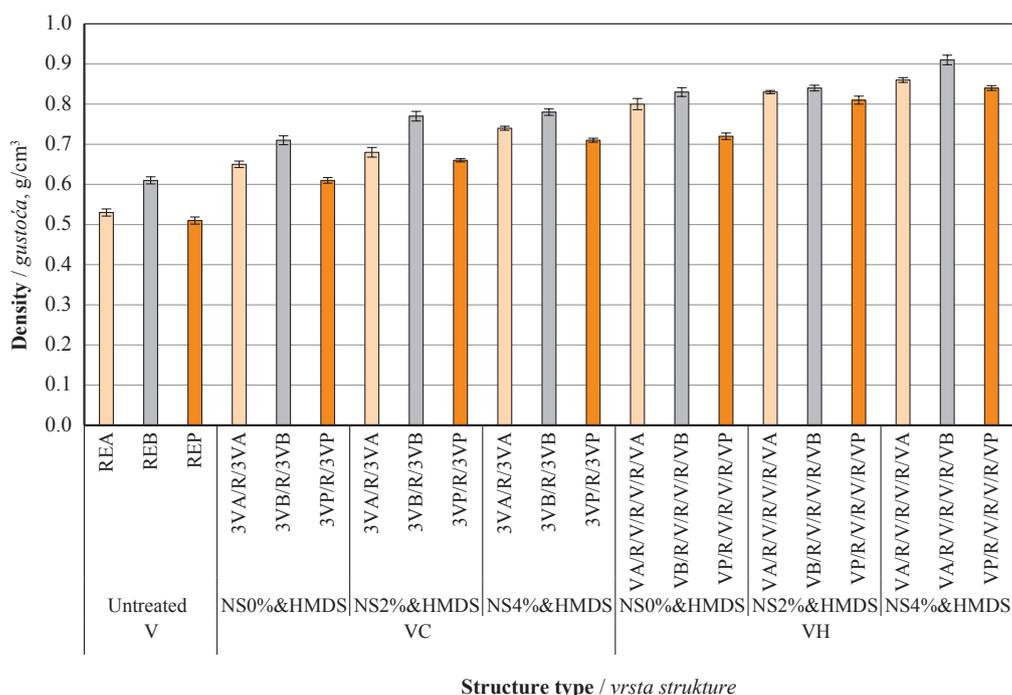


Figure 2 Density of composite plywood with different structures

Slika 2. Gustoća kompozitne furnirske ploče s različitim strukturama

Table 4 Physical-mechanical properties (Mean (\pm SD)) of plywood used**Tablica 4.** Fizičko-mehanička svojstva (srednja vrijednost, \pm SD) korištene furnirske ploče

| Type of plywood panels (RE) <i>Vrsta furnirske ploče (RE)</i> | Properties / Svojstva | | | | | | | | | |
|--|------------------------------|------------------------|------------------------|------------------------|------------------------|------------------------|---------------------------|--------------------------------|------------------------|------------------------|
| | <i>D</i> , g/cm ³ | <i>WU</i> , % | | <i>TS</i> , % | | <i>FS</i> , MPa | <i>FM</i> , MPa | <i>IBS</i> , kJ/m ² | <i>HS</i> , MPa | |
| | | 2 h | 24 h | 2 h | 24 h | | | | 5.5 mm | 7 mm |
| Alder <i>johovina</i> | 0.53 (\pm 0.008) | 37.61 (\pm 0.21) | 56.26 (\pm 1.25) | 11.75 (\pm 0.14) | 13.65 (\pm 0.30) | 76.80 (\pm 1.20) | 7933.33 (\pm 26.58) | 53.20 (\pm 0.29) | 46.58 (\pm 0.32) | 56.31 (\pm 0.20) |
| Beech <i>bukovina</i> | 0.61 (\pm 0.008) | 26.57 (\pm 0.32) | 47.65 (\pm 0.30) | 12.34 (\pm 0.17) | 16.69 (\pm 0.26) | 87.71 (\pm 1.62) | 9666.66 (\pm 22.50) | 64.73 (\pm 0.22) | 65.65 (\pm 0.32) | 76.53 (\pm 0.29) |
| Poplar <i>topolovina</i> | 0.51 (\pm 0.008) | 33.35 (\pm 0.32) | 51.75 (\pm 0.15) | 9.83 (\pm 0.15) | 12.59 (\pm 0.31) | 67.69 (\pm 1.76) | 7858.33 (\pm 23.16) | 46.39 (\pm 0.34) | 42.64 (\pm 0.26) | 51.52 (\pm 0.22) |

followed by alder (0.53 g/cm³) and poplar (0.51 g/cm³) (Table 4). The effect of the amount of nano-SiO₂, and the arrangement or alternation of the rubber layer and the wood layer on the density is shown in Figure 2.

As seen in Figure 2, the lowest density of plywood/rubber samples in three layers of beech, alder, and poplar is related to wooden layers with 0 wt% nano-SiO₂ content and rubber core layer, while the highest density of plywood/rubber samples were observed in three layers of beech, alder, and fir, corresponding to wooden layers with 4 wt% nano-SiO₂ content and side layers of rubber. The density of rubber ranges from 0.9 g/cm³ to 1 g/cm³ (Cheremisnoff, 2023). High density (2.6 g/cm³) and proper permeability of nano-SiO₂ increase the specific surface area (specific weight) of the produced composites (Mahzan *et al.*, 2010). The high density of mineral nanoparticles could be attributed to their high atomic level since a large number of atoms and the regular arrangement of mineral nanoparticles generally improve the physical characteristics (density) of the composite (Savov *et al.*, 2023). As can be seen, the density of the beechwood layer is higher than that of alder and poplar. The higher density of the beech layer compared to the alder and poplar layer can be related to its high density and low porosity (Ashori *et al.*, 2015; Toksoy *et al.*, 2006; Ilkay and Mengelöglu, 2022). So, the greater the porosity of the wood layer, the weaker the wood, leading to the decrease of the surface layer (Rodríguez *et al.*, 2014). Rubber as a polymer filler affects the density of composites. It seems that the low porosity, small molecular structure (macromolecular structure), and compactness of rubber particles can improve the specific mass (specific weight) of composites (Jun *et al.*, 2008; Xu and Li, 2012). The density of wood layers is around 0.40-0.68 g/cm³. Therefore, as the weight percentage of rubber powder increases, the density and porosity of the composites increase and decrease, respectively (Jun *et al.*, 2008).

3.1.2 Water uptake (WU)

3.1.2.1. Upijanje vode (WU)

The results obtained from the ANOVA indicated that, with the increase in the amount of nano-SiO₂ (4

wt%), the amount of water uptake increased significantly in 2 and 24 h (statistical confidence level 99 %; attached Table 1). Also, Duncan's test placed this state of combined treatment in group C with the highest amount of water uptake at 2 and 24 h compared to plywood/rubber samples. But in general comparison, the highest amount of water uptake in 2 and 24 h was observed in the control plywood samples, and Duncan's grouping placed this condition in group A (Supplement Table 2 and 3). The interaction of nano-SiO₂ content, arrangement, or rotation of rubber layer and wood layer on the water uptake (2 and 24 h) of wood and rubber layered composites is shown in Figure 3.

Water uptake of the pure plywood reflects the density. As the water uptake is expressed in percentages, it appears that wood with a lower density absorbs more water. However, although the plywood uptakes the same amount of water expressed in mass, the relative uptake expressed in percentages will be higher for the plywood with lower density. This phenomenon could explain the differences in water uptake between various wood-rubber composites. As expected, the composites that contain more rubber (V/R/V/R/V/R/V) uptake less water during immersion than the pure plywood. This phenomenon can be explained by the presence of hydrophobic rubber, the higher density of these composites, and the thickness of the exposed wood layer. For example, the outer layer of the composite 3V/R/3V is much thicker than the composite V/R/V/R/V/R/V; thus, the thicker outer layer can absorb more water during immersion than the thinner outer layer. In addition, the water uptake was influenced by the presence of the nano-SiO₂. Regardless of the wood type and the composition type, the best performance was determined by composites that contain 2 wt% of nano-SiO₂. The best combination was usually associated with beech wood. However, as mentioned, this cannot only be ascribed to better performance, but could also be the result of the differences in density. The results showed that nano-SiO₂ increases water uptake of the composite plywood. Since the surface of the nano-SiO₂ molecule has three functional groups (hydroxy, hydrogen attached to hydroxy groups (OH), and siloxane

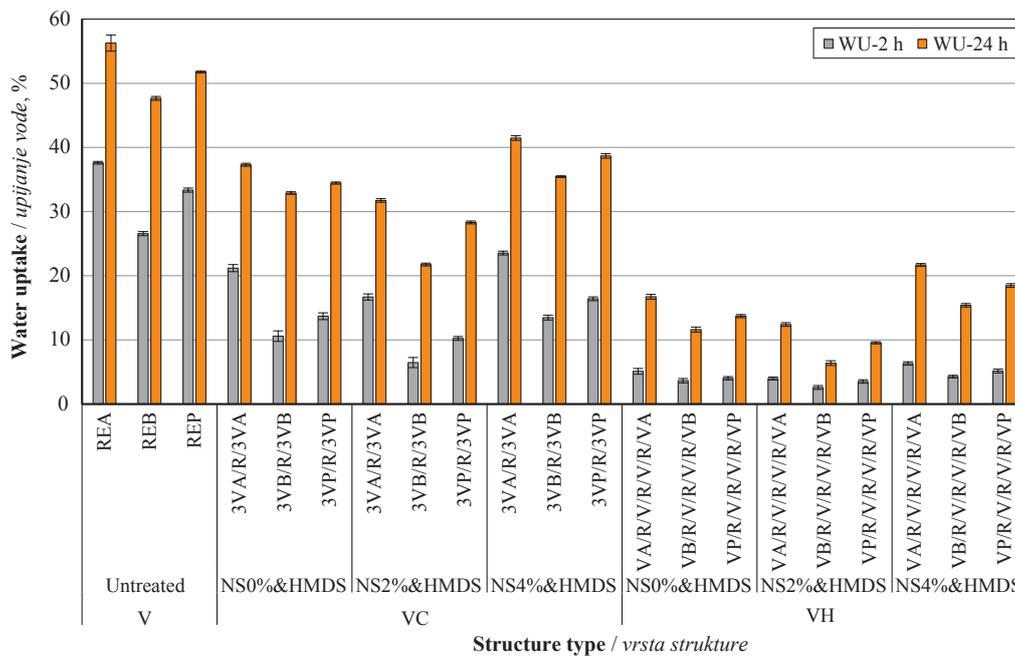


Figure 3 Water uptake (%) of composite plywood with different structures

Slika 3. Upijanje vode (%) kompozitne furnirske ploče s različitim strukturama

groups), silica particles are hydrophilic. In a sense, when the samples are placed in water, the nano-SiO₂ with very large lateral surfaces absorb water molecules and thus increase the amount of water uptake. Therefore, with the increase of nano-SiO₂ from 0 to 4 wt%, the amount of water uptake also increases, which can be due to the very high specific surface area of nano-SiO₂ particles and the hydrophilicity of their surface (Homkhiew *et al.*, 2015). Also, the results of the amount of water uptake in 24 h showed that the amount of water uptake of the alder layer is higher than that of poplar and beech. However, generally, the amount of water uptake of poplar, alder, and beech layer with rubber side layer and 0 and 2 wt% nano-SiO₂ was lower than that of the control plywood.

3.1.3 Thickness swelling (TS)

3.1.3.1. Debljinsko bubrenje (TS)

The results obtained from the ANOVA showed that, with the increase in the amount of nano-SiO₂ (4 wt%), the amount of thickness swelling in 2 and 24 h increased significantly (statistical confidence level 99%; attached Table 1). Also, Duncan's test placed this state of combined treatment in group C with the highest amount of thickness swelling at 2 and 24 h compared to plywood/rubber samples. But in general comparison, the highest amount of thickness swelling in 2 and 24 h was observed in the control plywood samples, and Duncan's grouping placed this condition in group A (Tables 4 and 5). Figure 4 depicts the expansion in the thickness of a wood and rubber layered composite after 2 and 24 h of immersion in water.

The visible growth in thickness agrees with the hydration information. Higher water uptakes result in higher thickness swelling. The highest thickness swelling was related to beech plywood (REB). This result is expected considering that beech is a wood species characterized by low dimensional stability (Wagenführ, 2014). The presence of rubber (R) has a positive effect on thickness swelling. First, because rubber limits water uptake, and second, because rubber absorbs part of the dimensional changes. However, there was an insignificant difference between the two respective types of composites. Both composites, 3V/R/3V and V/R/V/R/V/R/V, perform comparably. Similarly, as reported for water uptake, nano-SiO₂ has the most positive effect on a concentration of 2 wt%. Higher concentrations of nano-SiO₂ adversely affected the thickness swelling, which can be associated with higher water uptake. By increasing the amount of nano-SiO₂ up to 2 wt% level, some empty spaces and capillary tubes are occupied, which could result in a sharp reduction of these openings and make it difficult for water molecules to reach the wood material. Therefore, increasing the amount of nano-SiO₂ causes fewer water molecules to be absorbed by the wood material, resulting in reduced thickness swelling after 2 and 24 h (Kariminejad *et al.*, 2022). Rubber is very effective in reducing water uptake and thickness swelling. The decrease in water uptake and thickness in the side layer of the rubber is a consequence of the increase in the amount of rubber per unit volume of these panels and the water repellency of this material. Applying rubber for constructing multi-structures also showed a decrease in water up-

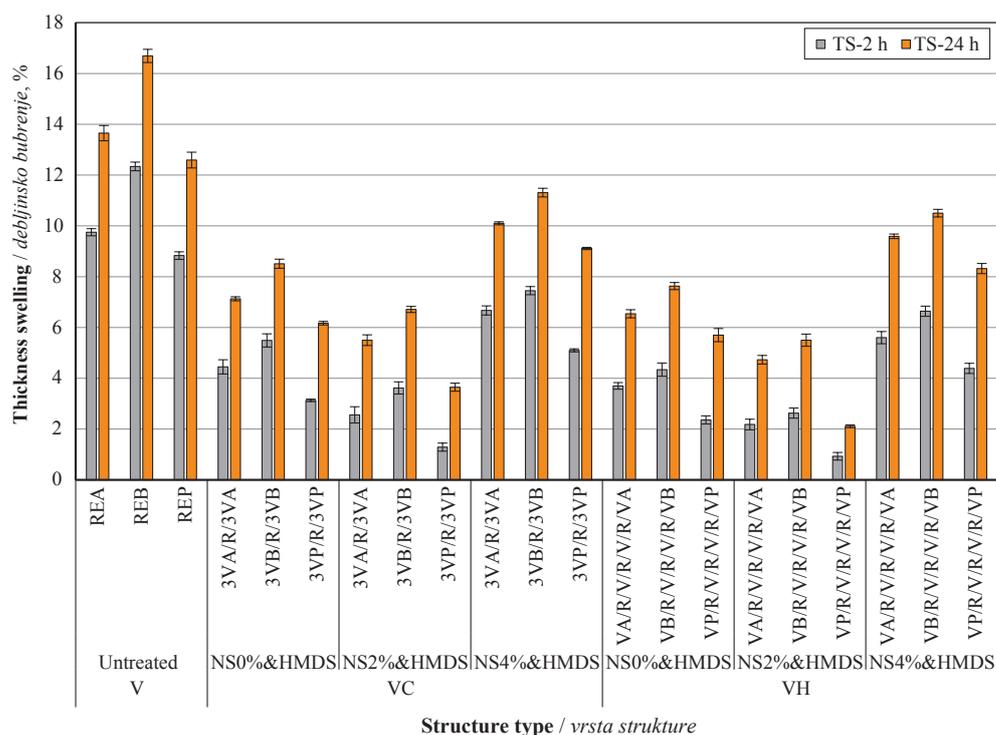


Figure 4 Thickness swelling (%) of composite plywood with different structures
Slika 4. Debljinsko bubrenje (%) kompozitne furnirske ploče s različitim strukturama

take and the thickness of these multi-structures with an increase in the ratio of rubber to wood (Mancel *et al.*, 2022; Ashori *et al.*, 2015; Ashori *et al.*, 2018). The low moisture absorption of polymeric materials (rubber) could lead to low moisture absorption of cellulose materials of composites. Therefore, by increasing the amount of rubber in the unit volume of multi-structures, the amount of water uptake and thickness swelling decreases (Ashori *et al.*, 2015; Ashori *et al.*, 2018).

3.2 Mechanical properties

3.2.1 Mehanička svojstva

3.2.1.1 Flexural strength (FS) and flexural modulus (FM)

3.2.1.1. Čvrstoća na savijanje (FS) i modul elastičnosti pri savijanju (FM)

The combined treatment of plywood/rubber samples did not significantly affect their modulus of elasticity (*MOE*) and modulus of rupture (*MOR*) at the 99 % confidence level, according to the ANOVA results (Table 1). Duncan's test indicated that the control plywood samples had the highest *MOE* and *MOR* among all groups, while the plywood/rubber samples with beech, alder and poplar layers, rubber side layer, and 0 wt% nano-SiO₂ content had the lowest *MOE* and *MOR* (Supplement Table 2 and 3). Adding rubber to the laminate flooring composite negatively impacted the flexural strength. Rubber is known for its elastic properties, which contributes to the decrease in *MOE* and *MOR*. As the wood is positioned in the

outer layer of the composite plywood, which has the most considerable effect on the bending strength, the presence of rubber insignificantly influenced the *MOE* and *MOR* mechanical properties. Regardless of the composition, the highest *MOE* and *MOR* were reported for the composites based on beech wood. This result is reasonable, as beech wood has much better mechanical properties than alder and poplar, predominantly due to higher density. In addition, the positive effect of the nano-SiO₂ is evident as well. *MOE* and *MOR* of the composites with higher concentrations of nano-SiO₂ are considerably better than those of the parallel composites without respective additives (Figure 5). The mechanical resistance of wood/plastic composites (*MOE* and *MOR*) is improved by increasing the content of nanoparticles (Deka and Maji, 2012). This occurs because an increase in nanoparticles leads to a greater dispersion of nanoparticles in the polymer matrix. As silica is spherical, it may be evenly distributed within the poly framework, where it will fill the voids between the wood fibers and the polymer matrix to produce a dense feel. As the composite density rises and its brittleness increases, the *MOE* and *MOR* rise as well (Moezipour *et al.*, 2013; Mohseni Tabar *et al.*, 2015). Some mechanical qualities of composites can be modified by adjusting the rubber-to-wood weight ratio. Both *MOE* and *MOR* go down as the rubber-to-wood weight ratio rises (Ashori *et al.*, 2015; Ashori *et al.*, 2018). Rubber, as a filler and cross-linking agent in composites, improves

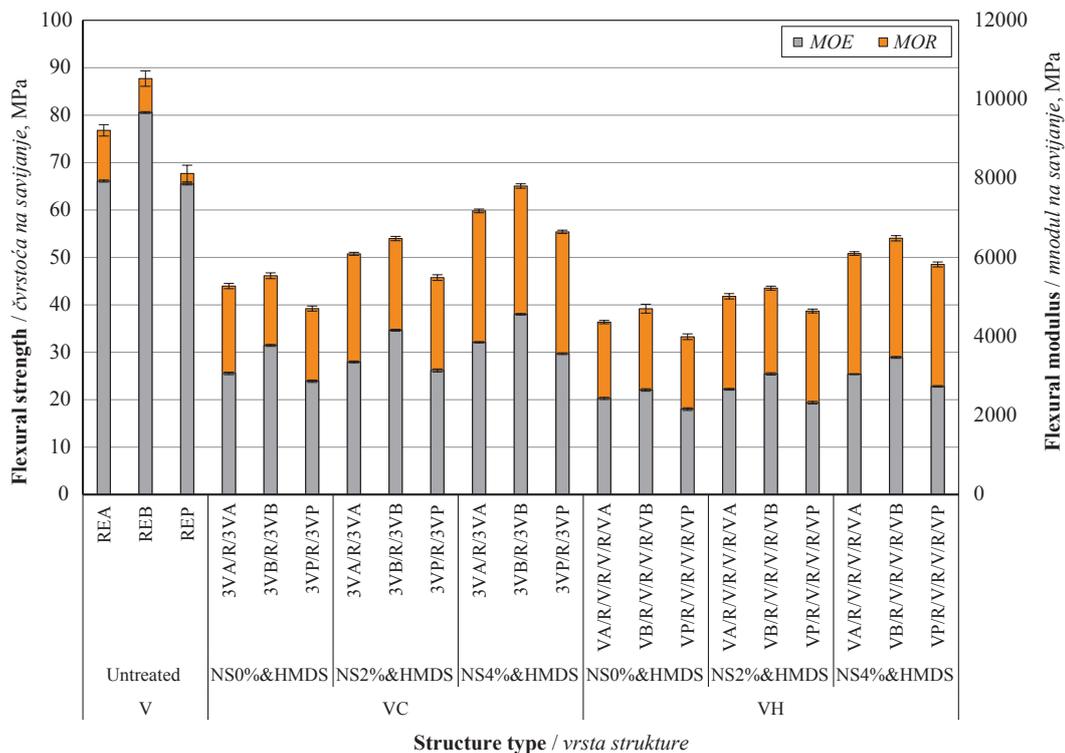


Figure 5 Flexural modulus and flexural strength of composite plywood with different structures
Slika 5. Modul elastičnosti pri savijanju i čvrstoća na savijanje kompozitne furnirske ploče s različitim strukturama

MOE and *MOR* (Ashori *et al.*, 2018; Jayadevan *et al.*, 2018; Cosnita *et al.*, 2014) despite its relatively modest amount compared to wood. The small molecular structure of rubber, or the small size of rubber particles, contributes to an increase in the specific surface area of rubber and a decrease in mechanical resistance (*MOE* and *MOR*) of composites (Ashori *et al.*, 2015).

3.2.2 Impact bending strength (*IBS*)

3.2.2.1 Udarne čvrstoća (*IBS*)

The ANOVA results showed that the combined treatment of plywood/rubber samples did not have a significant effect on their impact bending strength (*IBS*) at 99 % confidence level (Supplement Table 1). Duncan's test revealed that the control plywood samples had the highest *IBS* among all groups, while the plywood/rubber samples with beech, alder, and poplar layers, 4 wt% nano-SiO₂, and rubber side layer had the lowest *IBS* (Supplement Table 2 and 3). The results of the *IBS* are in line with the bending tests. Among plywood samples, the highest *IBS* was determined for beech plywood. This is predominately the result of the beech higher density compared to the other two hardwood species. Consequently, all composites that were based on beech wood had higher *IBS* in comparison to other composites based on poplar and alder. In addition, the composites with structure 3V/R/3V perform better than the ones with structure V/R/V/R/V/R/V, which is likely due to the fact that the outer layer of the

composites 3V/R/3V was more rigid than the other one. In addition, it can be seen that the addition of the additive nano-SiO₂ reduces the *IBS* (Figure 6). With the increase of nanoparticles, the impact resistance decreases (Nourbakhsh and Ashori, 2009; Yao *et al.*, 2015). In fact, nano-SiO₂ particles tend to attract each other because of their hydrophilic hydroxyl groups. In other words, nano-SiO₂ particles have high surface energy because hydroxyl groups can cause accumulation on the surface of the polymer by generating hydrogen bonds, leading to a decreased impact resistance (Elbarbary, 2022).

3.2.3 Hardness strength

3.2.3.1 Otpornost na zasijecanje

The ANOVA results showed that the combined treatment of plywood/rubber samples did not have a significant effect on their hardness strength at the 99 % confidence level (Supplement Table 1). Duncan's test revealed that the control plywood samples had the highest hardness strength among all groups, while the plywood/rubber samples with beech, alder and poplar layers, 4 wt% nano-SiO₂, and rubber side layer had the lowest hardness strength (Supplement Tables 2 and 3). Surface hardness is predominately affected by the hardness of the outer layer. Beech was the hardest among all tested wood species. Hence, the beech-rubber composites performed better than the ones based on poplar- and alder-rubber composites. As the outer layer of the rubber-wood composites, 3V/R/3V was

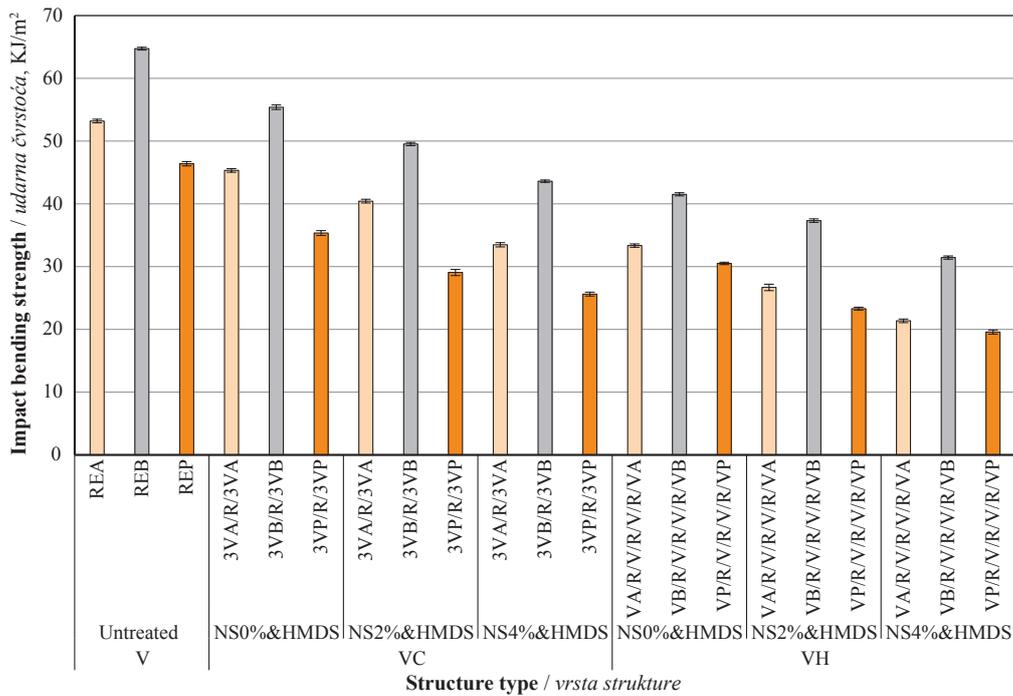


Figure 6 Impact bending strength of composite plywood with different structures
Slika 6. Udarna čvrstoća kompozitne furnirske ploče s različitim strukturama

thicker than V/R/V/R/V/R/V, resulting in higher hardness. The outer layer of the composite V/R/V/R/V/R/V was too thin to resist the penetration of the ball. The presence of the additive nano-SiO₂ has a similar effect as that of water. Middle concentration (2 wt%) has a positive effect, while the highest concentration of na-

no-SiO₂ did not contribute to the hardness (Figure 7). The higher specific surface ratio of rubber to wood reduces the adhesion between wood and glue; therefore, in addition to reducing the surface adhesion between wood layers, the mechanical characteristics (impact and hardness strength) of the composite are also re-

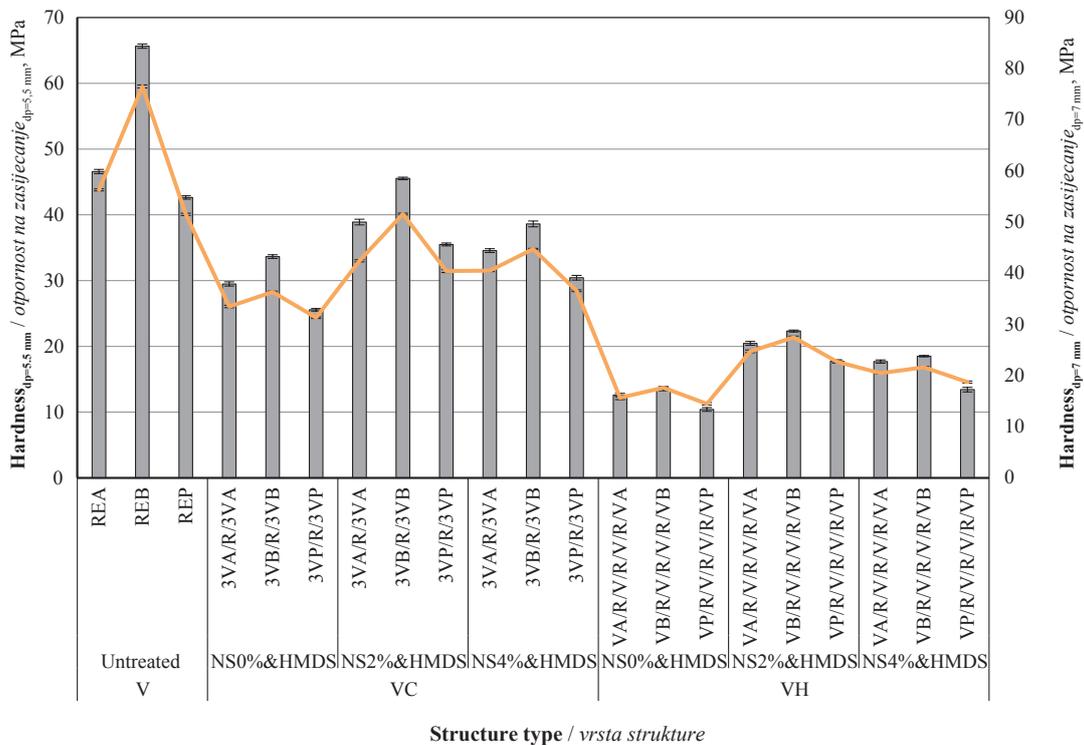


Figure 7 Surface hardness strength of composite plywood with different structures
Slika 7. Otpornost na zasijecanje kompozitne furnirske ploče s različitim strukturama

duced (Ashori *et al.*, 2015). The hardness strength of the composite is different in different penetration depths, so the hardness level increases with the increase of the penetration depth of the bullet (Taghiyari and Norton, 2014). The increase in hardness with a higher penetration depth compared to a lower penetration depth can be related to the greater penetration of the diameter of the bullet to the surface and finally the higher strength of the surface of the composite (Taghiyari and Norton, 2014).

4 CONCLUSIONS

4. ZAKLJUČAK

The objective of this study was to use recycled wood and rubber in the production of soft and strong composite flooring and also to investigate the physical-mechanical properties of the resulting multi-layered flooring. The results of this investigation showed that the three factors examined in this research, including the amount of modified nano-SiO₂, the wood layer, and the frequency or arrangement of the rubber layer, have a substantial effect on the physical and mechanical resistances (*D*, *WU* and *TS* in 2 and 24 h, *MOR* and *MOE*, *IBS*, and *HS*). In investigating the effect of the modified nano-SiO₂ loading, it was found that the density of the composites has increased. The amount of water uptake and thickness swelling after 2 and 24 hours of immersion in water showed that nano-SiO₂ did not have a meaningful effect on reducing water uptake and thickness swelling. Nano-SiO₂ had an effect on the improvement of *MOE*, *MOR* and *HS*, but it did not have a decisive effect on the improvement of *IBS*. The density of the wood layer had a great effect on the physical and mechanical resistance. So the wood layer with higher density enhanced the physical and mechanical resistance compared to the wood layer with lower density. However, in thickness swelling, wood layers of high density had more swelling; in other words, the thickness swelling of heavier wood layers was more pronounced than that of wood layers of low density. The arrangement of the rubber layer had a significant effect on increasing the physical properties, but it did not have a significant effect on the mechanical properties. Finally, it can be concluded that the use of modified nano-SiO₂ has the ability to improve some physical (density) and mechanical properties (*MOR*, *MOE*, and *HS*), and it can be used to improve the properties of filling materials such as elastic, plastic and other polymer materials.

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SUPPLEMENT – DODATAK

Table 1 ANOVA results for *D*, *WU*, *TS*, *FS*, *FM*, *IBS* and *HS*
Tablica 1. Rezultati ANOVA analize za *D*, *WU*, *TS*, *FS*, *FM*, *IBS* i *HS*

| Factor | | Property / Svojstvo | | | | | | | | | |
|---|----------------------------------|---------------------|-------------------|--------------------|------------------|------------------|------------------|-------------------|--------------------------------|--------------------|--------------------|
| | | <i>D</i> | <i>WU</i> | | <i>TS</i> | | <i>FS</i> , MPa | <i>FM</i> , MPa | <i>IBS</i> , kJ/m ² | <i>HS</i> | |
| | | | 2 h | 24 h | 2 h | 24 h | | | | 5.5 mm | 7 mm |
| Type of veneer <i>vrsta furnira</i> | <i>F</i> value <i>P</i> Value | 738.54 0.000 | 1751.81 0.000 | 4562.09 0.000 | 983.17 0.000 | 2250.49 0.000 | 1521.95 0.000 | 8887.01 0.000 | 22130.00 0.000 | 4465.45 0.000 | 6237.64 0.000 |
| SiO ₂ content <i>sadržaj SiO₂</i> | <i>F</i> value <i>P</i> Value | 794.86 0.000 | 878.43 0.000 | 12020.93 0.000 | 2954.74 0.000 | 9312.61 0.000 | 8361.89 0.000 | 6720.87 0.000 | 10549.14 0.000 | 8897.46 0.000 | 18126.34 0.000 |
| Arrangement of layer rubber <i>raspored slojeva gume</i> | <i>F</i> value <i>P</i> Value | 5073.97 0.000 | 15458.97 0.000 | 131792.98 0.000 | 374.46 0.000 | 749.37 0.000 | 6542.08 0.000 | 29378.71 0.000 | 27435.56 0.000 | 106642.41 0.000 | 199758.87 0.000 |
| Interaction <i>interakcija</i> | <i>F</i> value <i>P</i> Value | 25.68 0.000 | 1.526 0.201 | 72.78 0.000 | 3.798 0.007 | 6.792 0.000 | 8.54 0.000 | 15.38 0.000 | 30.78 0.000 | 18.61 0.000 | 145.25 0.000 |

Table 2 Duncan’s test results for the effect of wood veneer on studied properties
Tablica 2. Rezultati Duncanova testa utjecaja furnira drva na ispitivana svojstva

| Type of veneer <i>Vrsta furnira</i> | Property / Svojstvo | | | | | | | | | |
|--|------------------------------|---------------|---------|---------------|--------|-----------------|-----------------|--------------------------------|-----------------|---------|
| | <i>D</i> , g/cm ³ | <i>WU</i> , % | | <i>TS</i> , % | | <i>FS</i> , MPa | <i>FM</i> , MPa | <i>IBS</i> , kJ/m ² | <i>HS</i> , MPa | |
| | | 2 h | 24 h | 2 h | 24 h | | | | 5.5 mm | 7 mm |
| Alder / <i>johovina</i> | 0.76 B | 12.80 A | 26.88 A | 4.19 B | 7.27 B | 47.25 B | 3069.32 B | 33.44 B | 25.62 B | 29.56 B |
| Beech / <i>bukovina</i> | 0.81 A | 6.84 C | 20.58 C | 5.03 A | 8.36 A | 50.32 A | 3610.62 A | 43.14 A | 28.69 A | 33.23 A |
| Poplar / <i>topolovina</i> | 0.72 C | 8.84 B | 23.86 B | 2.87 C | 5.84 C | 43.48 C | 2799.48 C | 27.24 C | 22.18 C | 27.38 C |

Table 3 Duncan’s test results for the effect of nano-SiO₂ on studied properties
Tablica 3. Rezultati Duncanova testa utjecaja nano-SiO₂ na ispitivana svojstva

| Nano-SiO ₂ content, wt% <i>Sadržaj nano-SiO₂</i> <i>tež%</i> | Property / Svojstvo | | | | | | | | | |
|---|------------------------------|---------------|---------|---------------|--------|-----------------|-----------------|--------------------------------|-----------------|---------|
| | <i>D</i> , g/cm ³ | <i>WU</i> , % | | <i>TS</i> , % | | <i>FS</i> , MPa | <i>FM</i> , MPa | <i>IBS</i> , kJ/m ² | <i>HS</i> , MPa | |
| | | 2 h | 24 h | 2 h | 24 h | | | | 5.5 mm | 7 mm |
| NS0 | 0.72 C | 9.70 B | 24.45 B | 3.91 B | 6.95 B | 39.69 C | 2824.70C | 40.24 A | 20.87 C | 24.83 C |
| NS2 | 0.76 B | 7.25 C | 18.36 C | 2.20 C | 4.70 C | 45.75 B | 3115.67 B | 34.39 B | 30.08 A | 34.89 A |
| NS4 | 0.80 A | 11.53 A | 28.52 A | 5.97 A | 9.82 A | 55.61 A | 3539.06 A | 29.18 C | 25.54 B | 30.45 B |

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