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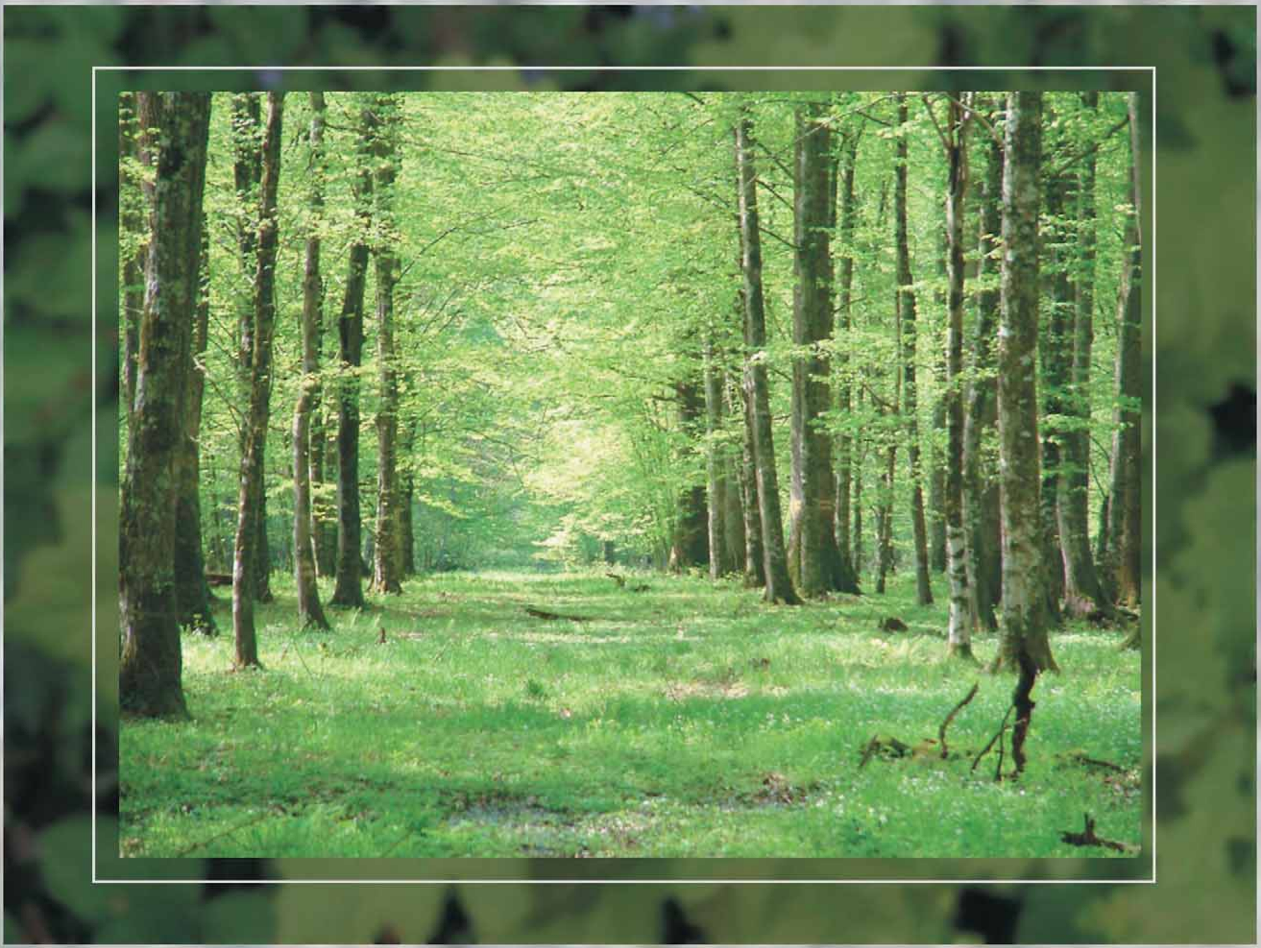









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# DRVNA INDUSTRIJA

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*Sezgin Koray Gülsoy*[1], *Saffet Uysal*[1]

# Effects of Recycled Fiber Fines on Hand Sheet Properties of Different Unbeaten and Beaten Pulps

## Utjecaj finih recikliranih vlakana na svojstva ručno izrađenog papira od nemljevene i mljevene celuloze

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**ABSTRACT •** *In this study, 5, 10, and 15 % secondary fines of recycled pulp were added to unbeaten and beaten (28 ºSR) samples of recycled pulp, Turkish Calabrian pine (Pinus brutia Ten.) kraft pulp, and European aspen (Populus tremula L.) kraft pulp. The effects of the addition of fiber fines on hand sheets properties were evaluated. The addition of fiber fines to the unbeaten pulps improved the strength properties of hand sheets. The roughness of hand sheets was also decreased with the addition of fines. When fiber fines were added to the beaten pulps, the type of pulp strongly affected the paper strength properties. The strength properties of beaten pulps of Turkish Calabrian pine and European aspen were decreased with the addition of fiber fines, while the strength properties of beaten pulps of recycled pulp were increased. On the other hand, the air permeance of unbeaten and beaten samples was decreased with the addition of fiber fines. Consequently, the addition of fines to unbeaten and beaten pulps had a more pronounced effect on European aspen kraft pulp and recycled pulp than on Turkish Calabrian pine kraft pulp. Also, the strength of paper made of unbeaten recycled pulp with the addition of 15 % fines was higher than that of fines-free beaten recycled pulp.*

***Keywords:*** *Populus tremula L.; Pinus brutia Ten.; recycled fiber; kraft; fines; pulp and paper properties*

**SAŽETAK •** *U radu je opisano istraživanje u kojemu je mljevenoj i nemljevenoj (28 °SR) recikliranoj celulozi, kraft celulozi od brucijskog bora (Pinus brutia Ten.) i kraf celulozi od jasike (Populus tremula L.) dodano 5, 10 i 15 % sekundarnih finih vlakana reciklirane celuloze. Utvrđeni su učinci dodavanja finih vlakana na svojstva ručno izrađenih papira. Dodatak vlakana nemljevenoj celulozi pridonio je povećanju čvrstoće i smanjenju hrapavosti ručno izrađenih papira. Pri dodavanju vlakana mljevenoj celulozi velik je utjecaj na čvrstoću papira imala vrsta celuloze. Čvrstoća mljevene celuloze od brucijskog bora i jasike dodatkom vlakana se smanjila, dok se čvrstoća mljevene reciklirane celuloze povećala. Usto, dodatkom vlakana smanjena je propusnost zraka mljevenih i nemljevenih uzoraka. Posljedično, učinak dodavanja vlakana mljevenoj i nemljevenoj celulozi bio je veći za kraft celulozu od jasike i recikliranu celulozu nego za kraft celulozu od brucijskog bora. Također, čvrstoća papira od mljevene reciklirane celuloze s dodatkom 15 % vlakana bila je veća od čvrstoće papira od mljevene celuloze bez dodatka vlakana.*

***Ključne riječi:*** *Populus tremula L; Pinus brutia Ten; reciklirana vlakna; kraft; fina vlakna; svojstva celuloze i papira*

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## 1 INTRODUCTION
## 1. UVOD

The fines are defined as the cellulosic fiber fraction of a fiber suspension which can pass through a 200 mesh (200 wires per inch) screen with a 14.5 % open area and 76.2 μm diameter in Bauer McNett classifier (Meyers and Nanko, 2005). Therefore, the fines are sometimes named the P200 (Taipale, 2010). Also, the maximum and minimum fine length is detected as 0.2 mm and 0.072 mm, respectively (Meyers and Nanko, 2005). On the other hand, fines are extensively evaluated as all particles smaller than wood fibers present in the furnish (Krogerus *et al.,* 2002a). In accordance with this definition, fines may consist of various particles such as parts of wood cells, inorganic pigments and fillers, colloidal resins and latex, salt crystals, precipitates, deposits, etc. (Taipale, 2010).

Fines are generally categorized as primary and secondary fines. Primary fines are present in the pulp prior to refining, whereas secondary fines arise during refining. Primary fines consist mainly of ray cells, parenchyma cells and middle lamella lignin. (Bäckström *et al.,* 2008). Primary or secondary layers of cell wall are delaminated during refining, and they became detached from the fibers. So-formed secondary fines incline to be slender and flexible (Chen *et al.,* 2009).

Primary and secondary fines contribute differently to the mechanical properties of paper (Htun and de Ruvo, 1978). For a given fines content, the effect of secondary fines on strength properties of the pulp are more pronounced than those of primary fines (Hawes and Doshi, 1986; Bäckström *et al.,* 2008). Primary fines have a higher lignin content than kraft pulp fibers (Retulainen *et al.,* 2002). Secondary fines come mostly from the fiber surface during refining, and they have a higher lignin content compared to the fibers, but lower compared to the primary fines (Lindström and Nordmark, 1978). Secondary fines have about twice as much fibrils as primary fines (Krogerus *et al.,* 2002b). In chemical pulp, the cellulose and hemicellulose content of fines is higher than in the fiber fraction (Taipale, 2010). Also, the crystallinity of fines was lower compared to the long fiber fraction (Waterhouse and Omori, 1993).

The term *fines* is extensively used in the papermaking process, and fines have a significant influence on the behavior of the wet web and on all the properties of the final sheet (Bäckström *et al.,* 2008). Fines can help to fill in the voids between fibers in the paper structure. Fibrillar fines can bring fibers into closer contact with each other during the consolidation and drying of sheet (Johansson, 2008). In other words, fines increase inter-fiber bonding by acting as a bridge between fibers (Bäckström *et al.,* 2008; Retulainen *et al.,* 2002; Silveira *et al.,* 1996). If there are more bonds, the segments between bonds are shorter and there are fewer free loops (Retulainen *et al.,* 1993). Thus, a denser, stronger, and more uniform product is formed (Lin *et al.,* 2007). On the other hand, high levels of fines can make it more difficult to dewater the wet sheet (Liu *et al.,* 2001; Krogerus *et al.,* 2002b; Seth, 2003; Hubbe and Heitmann, 2007). The difficulty in dewatering produces a deceleration in the manufacturing process (Htun and de Ruvo, 1978; Taipale, 2010), causing an increase in the amount of energy required to dry the paper (Chen *et al.,* 2009).

The effect of fines on paper properties of mechanical pulps (Mohlin, 1977; Moss and Retulainen, 1997; Lu, 1999; Luukko and Paulapuro, 1999; Rundlöf, 2002; Vainio *et al.,* 2007; Asikainen *et al.,* 2010; Chen *et al.,* 2013; Moberg *et al.,* 2014) and chemical pulps (Kibblewhite,1972; Lobben, 1977; Htun and de Ruvo, 1978; Hartman, 1984; Przybysz and Czechowski, 1985; Retulainen *et al.,* 1993, 2002; Retulainen, 1997; Ferreira *et al.,* 2000; Krogerus *et al.,* 2002a; Taipale *et al.,* 2010) have been extensively studied. However, the influences of the addition of fines on the paper properties of the recycled pulps have seldom been reported. In general, fines (primary and secondary fines) of the recycled pulps are considered as undesirable for the strength properties, because the fines are produced from dried and hornified fibers. Mancebo and Krokoska (1985) noted that the fines of recycled pulp have a negative impact on paper strength as they act as filler material. The fines indicate the loss of bonding ability due to hornification, and it is claimed that the effect is irreversible even with refining. Also, Szwarcsztajn and Przybysz (1977) found that fines and fibers obtained from recycled paper hornified, causing the loss of the paper strength. Quite the contrary, Hawes and Doshi (1986) noted that fines of recycled paper are effective in increasing the strength of hand sheets made from recycled paper. The recycled fines act like virgin fines, so the removal of fines causes a reduction in the bonding index (Htun and de Ruvo, 1978; Klungness and Sanyer, 1981; Rushdan, 2005). Recently, the effect of fine content and quality on the recycled chemical pulps was studied by Lee *et al.,* (2011).

In this study, the effects of the addition of secondary fines to recycled pulp (5, 10, and 15 %) on hand sheet properties were evaluated. Three types of pulp were used, recycled pulp, Turkish Calabrian pine (*Pinus brutia* Ten.) kraft pulp, and European aspen (*Populus tremula* L.) kraft pulp, in order to determine the relationship between the addition of fines and the type of pulp. Also, two types of freeness level were used, unbeaten pulp and 28 ºSR pulp, in order to investigate the effect of fines on pulps at different freeness levels.

## 2 MATERIALS AND METHODS
## 2. MATERIJALI I METODE

Unbleached recycled pulps used in this study were obtained from Oyka pulp mill in Turkey. Turkish Calabrian pine and European aspen wood samples were obtained from Bartin province of Turkey. The wood samples were debarked and chipped to 3 cm × 1.5 cm × 0.5 cm size for kraft pulping. The air dried wood chips were stored in dry conditions. The kraft cooking conditions of both species are given in Table 1.

Cooking was carried out in a 15-L electrically heated laboratory cylindrical-type rotary digester. After

**Table 1** Kraft cooking conditions of both species
**Tablica 1.** Uvjeti kuhanja kraft celuloze od obje vrste drva

| **Tree species**<br>*Vrste drva* | **Active alkali**<br>*Aktivna lužina*<br>% | **Sulfidity**<br>*Sulfidnost*<br>% | **Cooking temp**<br>*Temperatura kuhanja,*<br>°C | **Cooking time to max. temp**<br>*Vrijeme kuhanja do najviše temperature*<br>min | **Cooking time at max. temp**<br>*Vrijeme kuhanja na najvišoj temperaturi*<br>min | **Liquor/ wood ratio**<br>*Omjer otapalo/ drvo* |
|---|---|---|---|---|---|---|
| Turkish Calabrian pine<br>*brucijski bor* | 18 | 25 | 170 | 90 | 75 | 4/1 |
| European aspen / *jasika* | 16 | 20 | 170 | 90 | 60 | 4/1 |

digestion, pulps were washed to remove black liquor and were disintegrated in a laboratory-type pulp mixer. Disintegrated pulps were screened using a Somerville-type pulp screen with 0.15-mm slotted plate (TAPPI T 275). Pulps were then beaten to 28 ºSR in a Valley Beater according to TAPPI T 200. Kappa number (TAPPI T 236), screened yield (TAPPI T 210), viscosity (SCAN-CM 15-62), and freeness of pulps (ISO 5267-1) were determined according to relevant methods.

The secondary fines were acquired by refining the recycled pulps in a Valley beater for 7 h with loading according to TAPPI T 200. The refined pulp was fractionated in a Bauer–McNett classifier (TAPPI T 233) into five fractions: R30, R50, R100, R200, and R300 mesh. Fines passing through a 200 mesh screen and retained by a 300 mesh were used as paper additive. The fiber morphology of the different pulps and secondary fines was determined with light microscope. Some pulp properties and fiber morphology of different pulps and fines are given in Table 2.

Different mixtures of unbeaten and beaten pulps of different fibers (Turkish Calabrian pine, European aspen, and recycled fiber) and secondary fines were used to prepare hand sheets and to determine the effect of fines on paper strength properties. Four levels of fines were used: 0, 5, 10, and 15 % of the dry weight of the fibers (Table 3). More than 15 % fines were not used due to difficulty in dewatering and drainage during papermaking. Freeness of fines added pulps (ISO 5267-1) was also determined. The hand sheets (70 g/m$^2$) made by a Rapid-Kothen Sheet Former (ISO 5269-2) were conditioned (TAPPI T 402). Tensile index (ISO 1924-3), burst index (TAPPI T 403), tear index (TAPPI T 414), roughness (ISO 8791-2), apparent density (TAPPI T 220), and air permeance (ISO 5636-3) of the hand sheets were also determined.

The data of hand sheet properties were subjected to analysis of variance (ANOVAs) and Duncan test at a 0.05 probability level. The same lower case letter in all figures denotes that the difference in the average values

**Table 2** Some pulp properties and fiber morphology of different pulps and fines
**Tablica 2.** Svojstva celuloze i morfologija vlakana od različitih vrsta celuloze i finih vlakana

| **Sample** / *Uzorak* | **Viscosity**<br>*Viskoznost*<br>cm$^3$/g | **Kappa number**<br>*Kappa broj* | **Fiber lengthmm**<br>*Duljina vlakana*<br>mm | **Fiber width**<br>*Širina vlakana*<br>µm | **Lumen width**<br>*Širina lumena*<br>µm | **Cell wall thickness**<br>*Debljina stanične stijenke*<br>µm |
|---|---|---|---|---|---|---|
| Turkish Calabrian pine kraft pulp<br>*kraft celuloza od brucijskog bora* | 888 | 31.4 | 2.90 | 54.50 | 28.50 | 13.00 |
| European aspen kraft pulp<br>*kraft celuloza od jasike* | 1121 | 32.5 | 1.18 | 24.25 | 10.50 | 6.88 |
| Recycled pulp / *reciklirana celuloza* | 580 | 31.2 | 1.66 | 28.00 | 10.75 | 8.63 |
| Fines / *fina vlakna* | - | - | 0.21 | 23.50 | 11.00 | 6.25 |

**Table 3** Fiber and fines mixing ratios used in this study
**Tablica 3.** Omjer vlakana i finih vlakana u smjesi rabljenoj u ovom istraživanju

| **Code**<br>*Oznaka* | **Mixing ratio** / *Omjer miješanja* |
|---|---|
| A0 | 100 % Turkish Calabrian pine kraft pulp / *100 % kraft celuloze od brucijskog bora* |
| A5 | 95 % Turkish Calabrian pine kraft pulp + 5 % fines / *95 % kraft celuloze od brucijskog bora + 5 % finih vlakana* |
| A10 | 90 % Turkish Calabrian pine kraft pulp + 10 % fines / *90 % kraft celuloze od brucijskog bora+ 10 % finih vlakana* |
| A15 | 85 % Turkish Calabrian pine kraft pulp + 15 % fines / *85 % kraft celuloze od brucijskog bora + 15 % finih vlakana* |
| B0 | 100 % European aspen kraft pulp / *100 % kraft celuloze od jasike* |
| B5 | 95 % European aspen kraft pulp + 5 % fines / *95 % kraft celuloze od jasike + 5 % finih vlakana* |
| B10 | 90 % European aspen kraft pulp + 10 % fines / *90 % kraft celuloze od jasike + 10 % finih vlakana* |
| B15 | 85 % European aspen kraft pulp + 15 % fines / *85 % kraft celuloze od jasike + 15 % finih vlakana* |
| C0 | 100 % recycled pulp / *100 % reciklirane celuloze* |
| C5 | 95 % recycled pulp + 5 % fines / *95 % reciklirane celuloze + 5 % finih vlakana* |
| C10 | 90 % recycled pulp + 10 % fines / *90 % reciklirane celuloze + 10 % finih vlakana* |
| C15 | 85 % recycled pulp + 15 % fines / *85 % reciklirane celuloze + 15 % finih vlakana* |

of properties among the compared groups was statistically insignificant.

## 3 RESULTS AND DISCUSSION
## 3. REZULTATI I RASPRAVA

### 3.1 Pulp freeness
### 3.1. Sposobnost odvodnjavanja celuloze

Drainage, an important parameter in the paper manufacturing, restricts the production efficiency of a paper machine (Norell *et al.,* 1999). Important factors influencing wetness end drainage are pulp composition, average fiber length, fiber length distribution, fines content, charge level on the stock, and degree of stock hydration (Paradis *et al.,* 2002). Fines have a detrimental effect on dewatering of the pulp suspension due to their high water holding capacity (Htun and de Ruvo, 1978; Hartman, 1984; Waterhouse and Omiri 1993; Liu *et al.,* 2001; Hubbe and Heitmann 2007; Chen *et al.,* 2009; Taipale *et al.,* 2010; Lee *et al.,* 2011; Lindqvist *et al.,* 2011). Lu (1999) noted that the pulp containing 40 % of fines drains 10 times more slowly than fines-free pulp. Kibblewhite (1972) reported that the pulp freeness strongly affected the quality and quantity of fines. Seth (2003) noted that pulp freeness drastically decreases with the addition of fines. The effect of the addition of fines on pulp freeness of unbeaten and beaten pulps is given in Table 4. The pulp freeness was decreased with increasing fines ratio.

### 3.2 Tensile index
### 3.2. Vlačni indeks

Tensile index is one of the basic strength properties of paper. The effect of the addition of fines on tensile index of the unbeaten and beaten pulps was more pronounced in European aspen kraft pulp and recycled pulp than in Turkish Calabrian pine kraft pulp (Figure 1). On the other hand, the effect of the addition of fines on tensile index of unbeaten pulps was more prominent than of beaten pulps. The effect was more pronounced at a higher level of addition. Lobben (1977) reported that the fines of chemical pulps had a considerable effect on strength properties depending on fiber type and freeness level of pulp. The effect of the addition of fines was greater for a eucalypt kraft pulp than for a pine kraft pulp. Also, the effects were more pronounced in unbeaten pulps of long fiber fraction.

**Table 4** The effect of fines addition on pulp freeness of unbeaten and beaten pulps

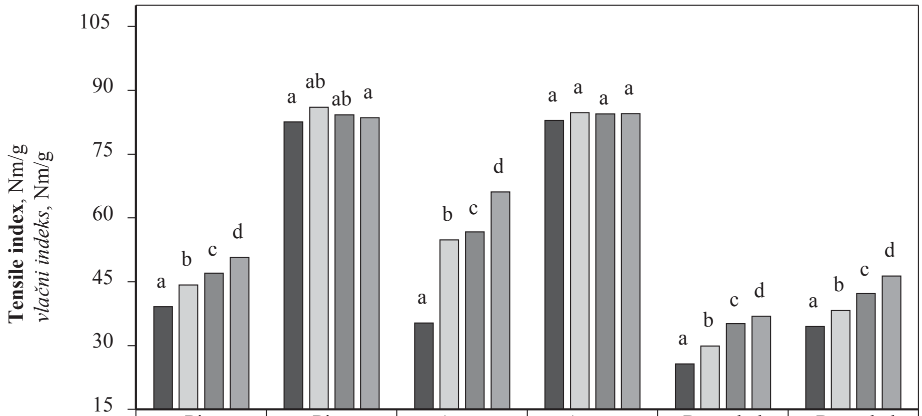
**Tablica 4.** Utjecaj dodatka finih vlakana na sposobnost odvodnjavanja mljevene i nemljevene celuloze

| Code *Oznaka* | **Unbeaten pulp** *Nemljevena celuloza* ºSR | **Beaten pulp** *Mljevena celuloza* ºSR |
|---|---|---|
| A0 | 13 | 28 |
| A5 | 15 | 41 |
| A10 | 16 | 50 |
| A15 | 27 | 59 |
| B0 | 13 | 28 |
| B5 | 17 | 38 |
| B10 | 22 | 45 |
| B15 | 28 | 51 |
| C0 | 20 | 28 |
| C5 | 25 | 35 |
| C10 | 34 | 43 |
| C15 | 37 | 55 |

As can be seen in Figure 1, the addition of 5, 10, and 15 % secondary fines to Turkish Calabrian pine unbeaten kraft pulps resulted in the increase in tensile index of 13.14 %, 20.22 %, and 29.68 %, respectively ($p<0.05$). The addition of 5, 10, and 15 % secondary fines to unbeaten European aspen kraft pulp resulted in the increase in tensile index of 55.22 %, 60.49 %, and 85.07 %, respectively. The addition of 5, 10, and 15 % secondary fines to recycled unbeaten pulps resulted in the increase in tensile index of 16.29 %, 36.80 %, and 43.49 %, respectively ($p<0.05$). Tensile index did not deteriorate with increasing addition of fines, and the more fines in hand sheet caused better bonding. These findings can be attributed to the increase in fiber-fiber bonding area due to the filling of inter-fiber gaps in the paper structure by fines. On the other hand, the addition of fines to fiber suspension results in the decreased average fiber length. However, tensile index of hand sheets did not decrease. This may be due to increasing fiber-fiber bonding thanks to the presence of fines, which overcompensated for the impaired tensile index caused by the decreasing average fiber length.

Previous studies revealed that the recycled fines act like virgin fines, causing an increase in the bonding index (Hawes and Doshi, 1986; Htun and de Ruvo, 1978; Klungness and Sanyer, 1981; Rushdan, 2005). In this study, the recycled fines did not act as a filler as found in other studies (Szwarcsztajn and Przybysz, 1977; Manchebo and Krokoska, 1985). Asikainen *et al.,* (2010) noted that tensile index increased with the addition of 10 % and 20 % primary fines to CTMP from 22.5 Nm/g to 26.8 Nm/g and 31.0 Nm/g, respectively. Retulainen (1997) reported that, by adding 3 % kraft fines to a kraft long fiber fraction, the tensile strength increases by 18 %. On the other hand, Lindqvist *et al.,* (2011) and Ferreira *et al.,* (2000) noted that the removal of fines resulted in decreased tensile index. The positive effect of the addition of fines on tensile index of hand sheets has been reported by several authors (Bäckström *et al.,* 2008; Retulainen, 1997; Mohlin, 1977; Hartman, 1984; Mancebo and Krokoska, 1985; Retulainen *et al.,* 1993; Waterhouse and Omiri, 1993; Waterhouse, 1994; Rundlöf, 2002; Vainio *et al.,* 2007; Taipale *et al.,* 2010; Zaytseva, 2010; Lee *et al.,* 2011).

In the beaten samples of Turkish Calabrian pine and European aspen kraft pulp, the addition of fines had not a statistically significant effect on tensile index of hand sheets ($p>0.05$) (Figure 1). The tensile index of recycled beaten pulps increased with the addition of 5, 10, and 15 % of secondary fines by 10.96 %, 22.47 %, and 34.47 %, respectively ($p<0.05$). These results can be attributed to better response to beating of virgin (flexible) fibers than recycled (stiff) fibers due to differences in their fiber morphology. Tensile index of



| | Pine unbeaten *nemljevena celuloza od bora* | Pine beaten *mljevena celuloza od bora* | Aspen unbeaten *nemljevena celuloza od jasena* | Aspen beaten *mljevena celuloza od jasena* | Recycled unbeaten *nemljevena celuloza od recikliranog drva* | Recycled beaten *mljevena celuloza od recikliranog drva* |
|---|---|---|---|---|---|---|
| ■ Control | 39.12 | 82.53 | 35.33 | 82.92 | 25.71 | 34.49 |
| □ 5% fines | 44.26 | 86.03 | 54.84 | 84.70 | 29.89 | 38.27 |
| ▨ 10% fines | 47.03 | 84.21 | 56.70 | 84.44 | 35.17 | 42.24 |
| ▧ 15% fines | 50.73 | 83.56 | 66.07 | 84.49 | 36.89 | 46.38 |

**Figure 1** Effect of fines addition on tensile index of hand sheets of unbeaten and beaten pulps
**Slika 1.** Utjecaj dodatka finih vlakana na vlačni indeks papira od nemljevene i mljevene celuloze

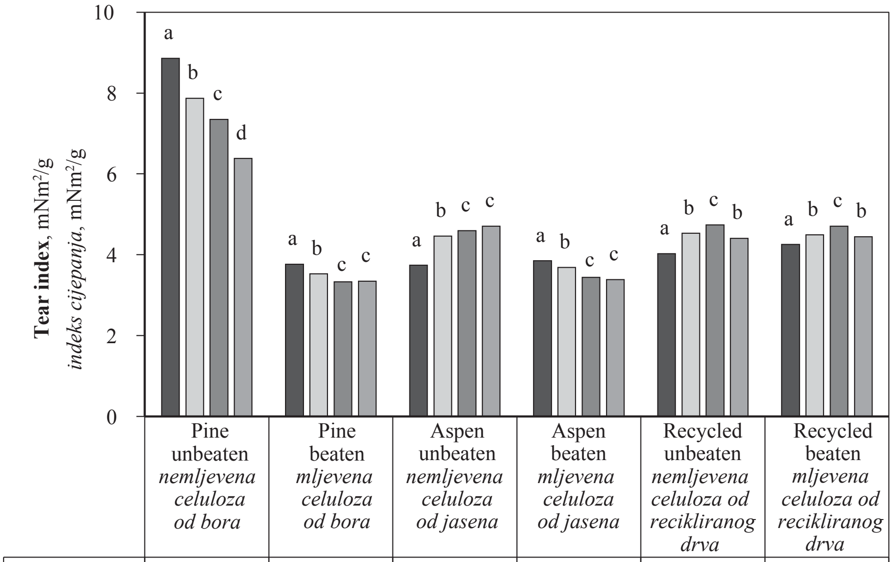
Turkish Calabrian pine and European aspen pulp samples was increased with beating to 28 ºSR by 110.97 % (from 39.12 Nm/g to 82.53 Nm/g) and 134.70 % (from 35.33 Nm/g to 82.92 Nm/g), respectively, while in recycled pulp samples this increase was only 34.15 % (from 25.71 Nm/g to 34.49 Nm/g). The lower tensile index increase in beaten recycled pulp was compensated with the addition of fines (Figure 1). On the other hand, tensile index of unbeaten recycled pulp (36.89 Nm/g), with the addition of 15 % fines, was higher than that of fines-free beaten recycled pulp (34.49 Nm/g).

### 3.3 Tear index
### 3.3. Indeks cijepanja

The effect of the addition of fines on tear index of hand sheet depends on the type of pulp and the amount of fines added. Tear index of Turkish Calabrian pine unbeaten kraft pulps decreased with the addition of 5, 10, and 15 % secondary fines by 11.17 %, 17.04 %, and 27.99 %, respectively ($p<0.05$). Quite the contrary, tear index of European black pine unbeaten kraft pulp and recycled unbeaten pulp increased with the addition of 5, 10, and 15 % secondary fines by 19.25 %, 22.99 %, 25.93 % and 12.40 %, 17.61 %, 9.42 %, respectively ($p<0.05$). These results showed that the addition of fines had a positive effect on tear index of hand sheets obtained from short fibers compared to long fibers (Figure 2). This result is consistent with previous work (Hawes and Doshi, 1986), which showed that tear index increased with the addition of 20 % secondary fines to recycled pulp from 8.7 mNm$^2$/g to 9.95 mNm$^2$/g. Ferreira *et al.,* (2000) noted that tear index increased from 9.5 mNm$^2$/g to 10.2 mNm$^2$/g when fines were removed from fiber suspension. The positive correlation between tear index and fines addition has also been reported by several authors (Mohlin, 1977; Hartman, 1984; Lee *et al.,* 2011). Quite the contrary, tear index decreased with the addition of fines (Waterhouse, 1994).

Tear index of Turkish Calabrian pine unbeaten kraft pulp decreased with the increasing addition of fines, while it increased for European aspen unbeaten kraft pulp and unbeaten recycled pulp (Figure 2). This result may be explained as follows. Tear index depends on several factors, including fiber length, wall thickness, inter-fiber bonding, fiber strength, etc. Fines contribute to tear index by increasing inter-fiber bonding due to their good flexibility and large surface area. Thus, tear index of short fiber European aspen and recycled pulp samples was increased with the addition of fines. As fines content increased, the average fiber length and wall thickness decreased. Also, the larger gaps between long fibers of Turkish Calabrian pine were not sufficiently filled by fines, resulting in lower inter-fiber bonding than samples without fines. Hence, tear index of Turkish Calabrian pine sample was decreased with the addition of fines.

In beaten pulps, tear index of Turkish Calabrian pine and European aspen kraft pulps decreased with the addition of 5, 10, and 15 % secondary fines by 6.37 %, 11.67 %, 11.14 % and 4.15 %, 10.65 %, 11.95 %, respectively ($p<0.05$) (Figure 2). Quite the contrary, the addition of 5, 10, and 15 % secondary fines to recycled unbeaten pulp resulted in the increase in tear index of 5.39 %, 10.56 %, and 4.46 %, respectively ($p<0.05$). These results showed that the effect of fines addition on tear index was more prominent than beating. For example, tear index of recycled unbeaten pulp was increased by beating up to 28 ºSR from 4.03 mNm$^2$/g to 4.26 mNm$^2$/g. However, with the addition of 15 % fines, tear



| | Pine unbeaten *nemljevena celuloza od bora* | Pine beaten *mljevena celuloza od bora* | Aspen unbeaten *nemljevena celuloza od jasena* | Aspen beaten *mljevena celuloza od jasena* | Recycled unbeaten *nemljevena celuloza od recikliranog drva* | Recycled beaten *mljevena celuloza od recikliranog drva* |
|---|---|---|---|---|---|---|
| ■ Control | 8.86 | 3.77 | 3.74 | 3.85 | 4.03 | 4.26 |
| □ 5% fines | 7.87 | 3.53 | 4.46 | 3.69 | 4.53 | 4.49 |
| ▩ 10% fines | 7.35 | 3.33 | 4.60 | 3.44 | 4.74 | 4.71 |
| ▨ 15% fines | 6.38 | 3.35 | 4.71 | 3.39 | 4.41 | 4.45 |

**Figure 2** Effect of fines addition on tear index of hand sheets of unbeaten and beaten pulps
**Slika 2.** Utjecaj dodatka finih vlakana na indeks cijepanja papira od nemljevene i mljevene celuloze

index of recycled unbeaten pulp was increased from 4.03 mNm$^2$/g to 4.41 mNm$^2$/g. On the other hand, when 15 % fines were added to unbeaten recycled pulp (4.41 mNm$^2$/g), tear index was higher than that of fines-free beaten recycled pulp (4.26 mNm$^2$/g).

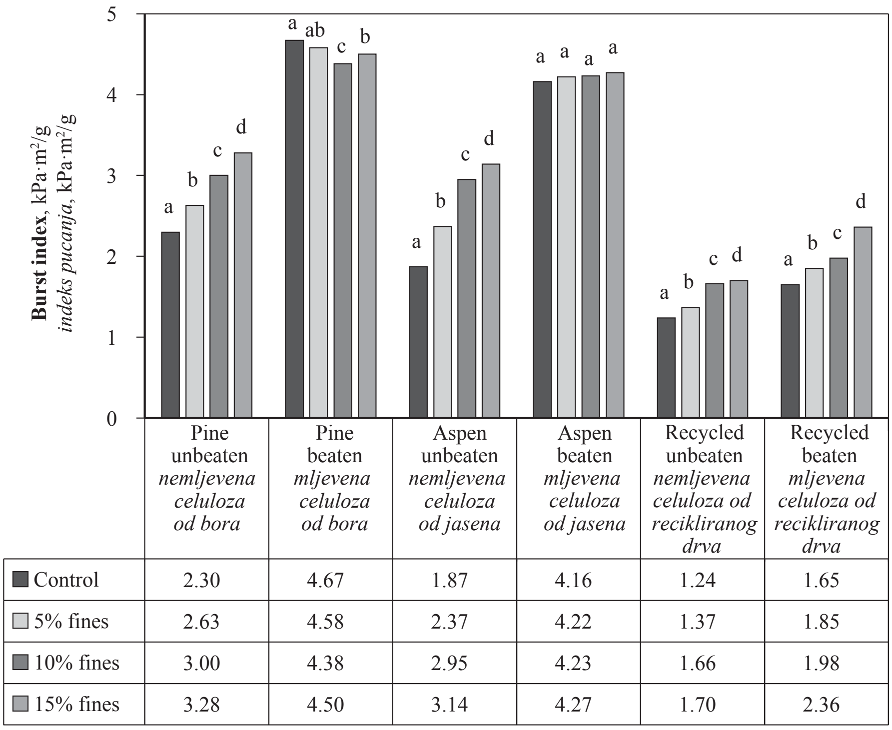
## 3.4 Burst index
### 3.4. Indeks pucanja

In unbeaten pulps of all types of pulp, the addition of fines caused an increase in burst index (Figure 3). The highest increase in burst index was determined in European aspen kraft pulp. The addition of 5, 10, and 15 % secondary fines to Turkish Calabrian pine kraft pulp resulted in the increase in burst index of 14.34 %, 30.43 %, and 42.60 %, respectively ($p<0.05$). The increase of burst index in European aspen kraft pulp and recycled pulp was found to be 26.73 %, 57.75 %, 67.91 % and 10.48 %, 33.87 %, 37.09 %, respectively ($p<0.05$). The burst index of hand sheets increased linearly with increasing proportion of fines (Figure 3). These findings can be ascribed to increasing inter-fiber bonding due to higher surface area of fines than fibers. The surface area of fines ranges from 10 to 50 m$^2$/g, while the surface area of fibers is around 1 m$^2$/g (Retulainen *et al.,* 1993).

Hawes and Doshi (1986) reported that burst index increased with the addition of 20 % secondary fines to recycled pulp from 1.39 kPam$^2$/g to 3.86 kPam$^2$/g. Ferreira *et al.*, (2000) noted that burst index decreased from 6.9 kPam$^2$/g to 5.0 kPam$^2$/g when fines were removed from fiber suspension. Other authors reported that burst index increases with the addition of fines (Mohlin, 1977; Htun and de Ruvo, 1978; Klungness and Sanyer, 1981; Hawes and Doshi, 1986; Waterhouse, 1994; Bäckström *et al.,* 2008).

In beaten pulps, the addition of 5, 10, and 15 % secondary fines to Turkish Calabrian pine kraft pulp resulted in burst index loss of 1.93 %, 6.21 %, and 3.64 %, respectively ($p<0.05$). Quite the contrary, the addition of secondary fines to European aspen kraft pulp resulted in a statistically insignificant increase of burst index, ($p>0.05$). On the other hand, an increase in burst index of 12.12 %, 20 %, and 43.03 %, respectively ($p<0.05$), was observed in recycled pulp with the addition of 5, 10, and 15 % secondary fines. In beaten pulps, the relationship between burst index and fines addition is drastically different depending on the type of pulp (Figure 3). These results can be explained by increased inter-fiber bonding that acts as a bridge of fines between hornified and stiff recycled fibers. Thus, the strength of paper made from recycled fibers with low bonding capacity increases with the addition of fines. Along with the beating, the decrease in fiber length and the improved fiber flexibility and plasticity increase the bonded area between fibers. Thus, the paper structure became more compact and burst index increased. Also, the mobility of fines may be lower in the wet sheet of collapsed, swollen, and externally fibrillated fibers compared to the more open structure in a wet sheet of unbeaten fibers (Hartman, 1984).

In the virgin pulp samples, freeness level of pulp had a more significant effect on burst index than the fines content. For example, burst index of Turkish Calabrian pine kraft pulp was increased with beating up to 28 °SR from 2.30 kPam$^2$/g to 4.67 kPam$^2$/g. However, burst index of Turkish Calabrian pine unbeaten kraft pulp was increased from 2.30 kPam$^2$/g to 3.28 kPam$^2$/g with the addition of 15 % fines. In the recycled pulp sample, the effect of freeness level of pulp and fines addition on burst index was similar. Burst index of recy-



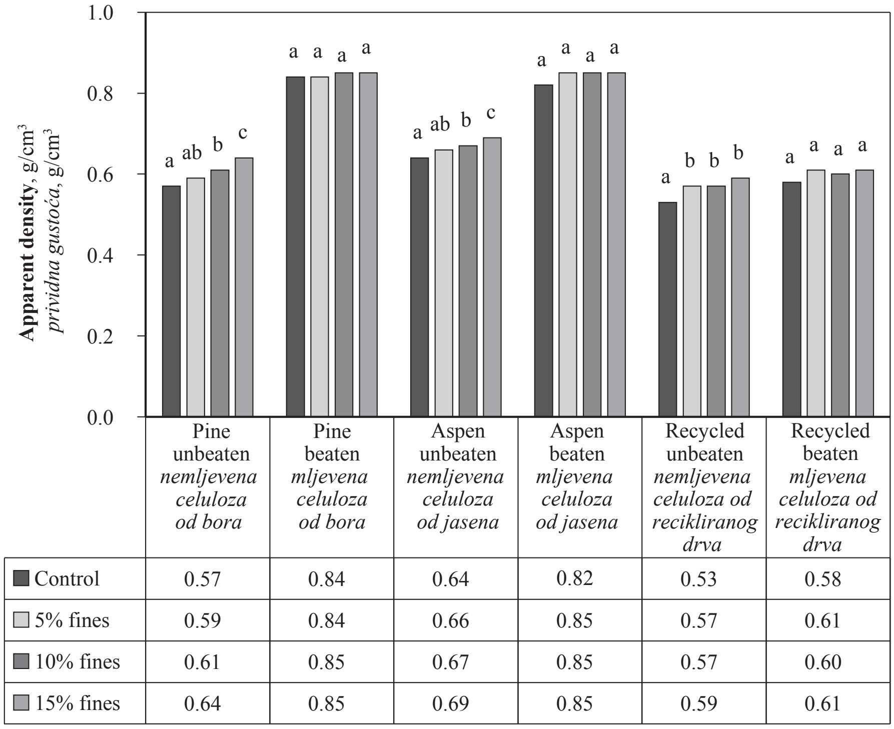
| | Pine unbeaten *nemljevena celuloza od bora* | Pine beaten *mljevena celuloza od bora* | Aspen unbeaten *nemljevena celuloza od jasena* | Aspen beaten *mljevena celuloza od jasena* | Recycled unbeaten *nemljevena celuloza od recikliranog drva* | Recycled beaten *mljevena celuloza od recikliranog drva* |
|---|---|---|---|---|---|---|
| ■ Control | 2.30 | 4.67 | 1.87 | 4.16 | 1.24 | 1.65 |
| □ 5% fines | 2.63 | 4.58 | 2.37 | 4.22 | 1.37 | 1.85 |
| ▩ 10% fines | 3.00 | 4.38 | 2.95 | 4.23 | 1.66 | 1.98 |
| ▨ 15% fines | 3.28 | 4.50 | 3.14 | 4.27 | 1.70 | 2.36 |

**Figure 3** Effect of fines addition on burst index of hand sheets of unbeaten and beaten pulps
**Slika 3.** Utjecaj dodatka finih vlakana na indeks pucanja papira od nemljevene i mljevene celuloze

cled pulp was increased with beating up to 28 ºSR from 1.24 kPam$^2$/g to 1.65 kPam$^2$/g. However, burst index of recycled unbeaten pulp was increased from 1.24 kPam$^2$/g to 1.70 kPam$^2$/g with the addition of 15 % fines.

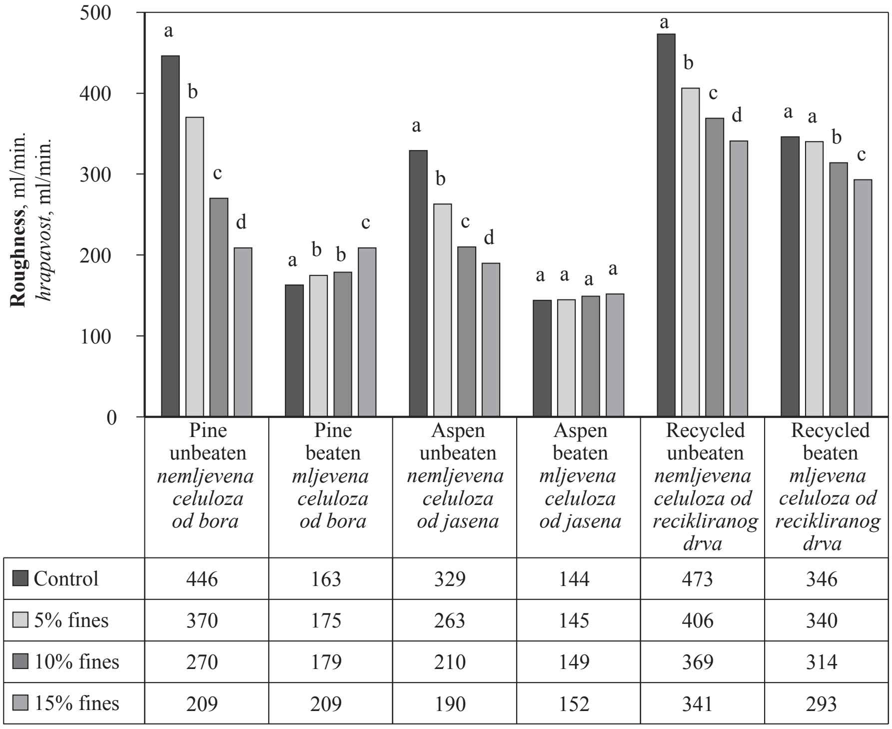
### 3.5 Apparent density
### 3.5. Prividna gustoća

As the addition of fines increased, the apparent density of the hand sheets increased linearly as shown in Figure 4. The highest increase in apparent density was determined in Turkish Calabrian pine kraft pulp. The addition of 5, 10, and 15 % secondary fines to Turkish Calabrian pine kraft pulp resulted in an increase in apparent density of 3.51 %, 7.02 %, and 12.28 %, respectively ($p<0.05$). An increase in apparent density of 3.13 %, 4.68 %, 7.81 % and 7.55 %, 7.55 %, 11.32 %, respectively ($p<0.05$), was determined in European aspen kraft pulp and recycled pulp. This is consistent



| | Pine unbeaten *nemljevena celuloza od bora* | Pine beaten *mljevena celuloza od bora* | Aspen unbeaten *nemljevena celuloza od jasena* | Aspen beaten *mljevena celuloza od jasena* | Recycled unbeaten *nemljevena celuloza od recikliranog drva* | Recycled beaten *mljevena celuloza od recikliranog drva* |
|---|---|---|---|---|---|---|
| ■ Control | 0.57 | 0.84 | 0.64 | 0.82 | 0.53 | 0.58 |
| □ 5% fines | 0.59 | 0.84 | 0.66 | 0.85 | 0.57 | 0.61 |
| ▩ 10% fines | 0.61 | 0.85 | 0.67 | 0.85 | 0.57 | 0.60 |
| ▨ 15% fines | 0.64 | 0.85 | 0.69 | 0.85 | 0.59 | 0.61 |

**Figure 4** Effect of fines addition on apparent density of hand sheets of unbeaten and beaten pulps
**Slika 4.** Utjecaj dodatka finih vlakana na prividnu gustoću papira od nemljevene i mljevene celuloze



| | Pine unbeaten *nemljevena celuloza od bora* | Pine beaten *mljevena celuloza od bora* | Aspen unbeaten *nemljevena celuloza od jasena* | Aspen beaten *mljevena celuloza od jasena* | Recycled unbeaten *nemljevena celuloza od recikliranog drva* | Recycled beaten *mljevena celuloza od recikliranog drva* |
|---|---|---|---|---|---|---|
| ■ Control | 446 | 163 | 329 | 144 | 473 | 346 |
| □ 5% fines | 370 | 175 | 263 | 145 | 406 | 340 |
| ▩ 10% fines | 270 | 179 | 210 | 149 | 369 | 314 |
| ▨ 15% fines | 209 | 209 | 190 | 152 | 341 | 293 |

**Figure 5** Effect of fines addition on roughness of hand sheets of unbeaten and beaten pulps
**Slika 5.** Utjecaj dodatka finih vlakana na hrapavost papira od nemljevene i mljevene celuloze

with previous work (Lu 1999), which showed that the sheet density increased with the addition of 20, 30, and 40 % fines to mechanical pulp from 0.372 g/cm$^3$ to 0.511 g/cm$^3$, 0.555 g/cm$^3$ and, 0.563 g/cm$^3$, respectively.

Fines cause an increase in sheet density by filling the voids between the fibers. In addition, Hawes and Doshi (1986) noted that the sheet density increased with the addition of 20 % secondary fines to recycled pulp from 0.549 g/cm$^3$ to 0.629 g/cm$^3$. On the other hand, Ferreira *et al.,* (2000) reported that sheet density decreased when fines were removed from fiber suspension from 0.767 g/cm$^3$ to 0.708 g/cm$^3$. The positive correlation between fine addition and sheet density has also been reported by other authors (Mohlin, 1977; Hartman, 1984; Hawes and Doshi, 1986; Waterhouse and Omiri, 1993; Lu, 1999; Bäckström *et al.,* 2008; Lee *et al.,* 2011; Moberg *et al.,* 2014).

In beaten pulps of all types of pulp, the addition of fines had a statistically insignificant effect ($p$>0.05) on apparent density of hand sheets (Figure 4). These results indicated that the freeness level of pulp is more important for the apparent density than the content of fines. For example, apparent density of European aspen kraft pulp was increased with beating up to 28 ºSR from 0.64 g/cm$^3$ to 0.82 g/cm$^3$. Whereas, apparent density of European aspen kraft pulp with the addition of 15% fines was increased from 0.64 g/cm$^3$ to 0.69 g/cm$^3$.

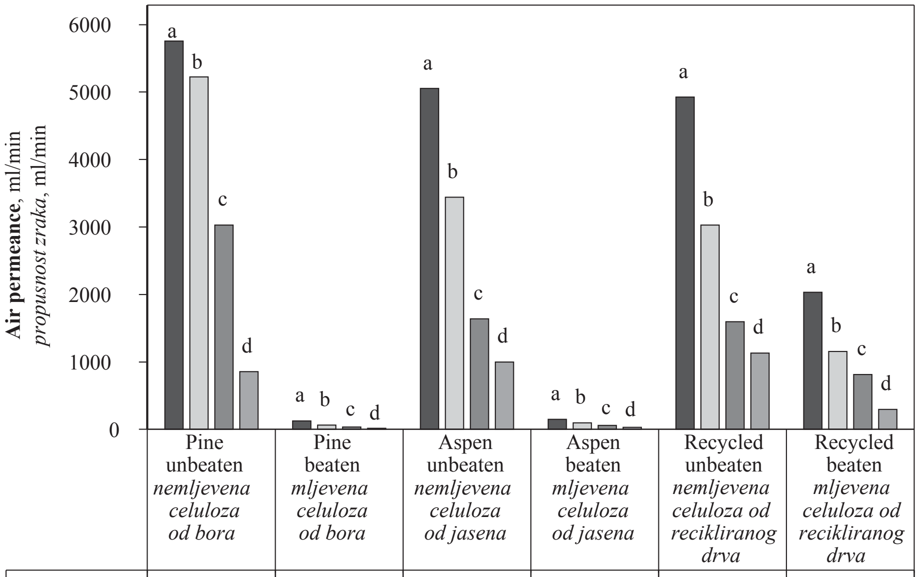
### 3.6 Roughness
### 3.6. Hrapavost

Hand sheet roughness was significantly affected by fines. In unbeaten pulps of all types of pulp, roughness of hand sheets decreased with the addition of fines (Figure 5). The effect of fines addition on roughness of Turkish Calabrian pine hand sheets was more pronounced than that of other pulps. In Turkish Calabrian pine unbeaten kraft pulp, the addition of 5, 10, and 15 % secondary fines resulted in a decrease in roughness of 17.04 %, 39.46 %, and 53.14 %, respectively ($p$<0.05). Roughness decrease in European aspen kraft pulp and recycled pulp was found to be 20.06 %, 36.17 %, 42.25 % and 14.16 %, 21.98 %, 27.90 %, respectively ($p$<0.05). These results can be explained by the fact that fines act as a filler material in the paper structure. Positive effect of fines on surface smoothness was also reported by Waterhouse and Omiri (1993) and Lu (1999).

In beaten pulps of Turkish Calabrian pine kraft pulp, hand sheet roughness increased with the addition of 5, 10, and 15 % secondary fines by 7.36 %, 9.82 %, and 28.22 %, respectively ($p$<0.05). Quite the contrary, the addition of 5, 10, and 15 % fines to recycled beaten pulp resulted in a roughness decrease of 1.73 %, 9.25 %, and 15.32 %, respectively ($p$<0.05). The effect of fines on hand sheet roughness of European aspen kraft pulp was statistically insignificant ($p$>0.05) (Figure 5).

### 3.7 Air permeance
### 3.7. Propusnost zraka

In unbeaten pulps of all types of pulp, air permeance of hand sheets decreased dramatically with increasing addition of fines (Figure 6). In Turkish Calabrian pines samples, the effect of fines on air permeance was more prominent than in other pulps. In Turkish Calabrian pine unbeaten kraft pulp, the addition of 5, 10, and 15 % fines caused an increase in air permeance of 9.24 %, 47.35 %, and 85.11 %, respectively ($p$<0.05). Air permeance of hand sheets of European aspen unbeaten kraft pulp and recycled unbeaten pulp decreased by 31.91 %, 67.54 %, 80.20 %, and



| | Pine unbeaten *nemljevena celuloza od bora* | Pine beaten *mljevena celuloza od bora* | Aspen unbeaten *nemljevena celuloza od jasena* | Aspen beaten *mljevena celuloza od jasena* | Recycled unbeaten *nemljevena celuloza od recikliranog drva* | Recycled beaten *mljevena celuloza od recikliranog drva* |
|---|---|---|---|---|---|---|
| ■ Control | 5756 | 126 | 5052 | 151 | 4925 | 2032 |
| □ 5% fines | 5224 | 62 | 3440 | 95 | 3028 | 1156 |
| ▩ 10% fines | 3030 | 36 | 1640 | 60 | 1597 | 815 |
| ▨ 15% fines | 857 | 17 | 999 | 32 | 1133 | 298 |

**Figure 6** Effect of fines addition on air permeance of hand sheets of unbeaten and beaten pulps
**Slika 6.** Utjecaj dodatka finih vlakana na propusnost zraka papira od nemljevene i mljevene celuloze

38.52 %, 67.57 %, 76.99 %, respectively ($p<0.05$), with the addition of 5, 10, and 15 % fines. Increasing the amount of fines in furnish also leads to decreased air permeance. Fines cause the decrease in air permeance by filling the voids of the fiber network. In other words, air permeance (Figure 6) tends to decrease with the addition of fines due to better conformity, reduction of spaces between fibers and increase of resistance to air flow (Yasumura *et al.,* 2012). Decreasing air permeance with the addition of fines can also be attributed to increasing apparent density with the addition of fines. At higher apparent densities, the pores in the sheet begin to close up due to better bonding. This led to the decrease in air permeance. Reduced air permeance retards moisture escape and slows the drying rate. Both factors contribute to decreased paper machine productivity (Seth, 2003).

Lu (1999) reported that air permeance decreased with the addition of 20, 30, and 40 % fines to mechanical pulp from 2538 ml/min. to 922 ml/min., 124 ml/min., and 52 ml/min. respectively. Asikainen *et al.,* (2010) noted that air resistance (Gurley method) increased with the addition of 10 % and 20 % primary fines to CTMP from 2.8 sec. to 7.7 sec. and 22.2 sec., respectively. On the other hand, Ferreira *et al.,* (2000) noted that air permeance decreased when fines were removed from fiber suspension from 54.5 ml/min. to 26 ml/min. Htun and de Ruvo (1978) reported that the removal of fines resulted in increased air permeability. The effect of fines on hand sheet air permeance has also been reported previously (Hartman, 1984; Hawes and Doshi, 1986; Waterhouse and Omiri, 1993; Lu, 1999; Seth, 2003; Hubbe and Heitmann, 2007; Lee *et al.,* 2011).

Regarding unbeaten pulp samples, air permeance of beaten pulps decreased with the addition of fines (Figure 6). There was a linear correlation between fines addition ratio and air permeance of hand sheets. Air permeance of beaten kraft pulps of Turkish Calabrian pine and European aspen decreased by 50.79 %, 71.43 %, 86.51 %, and 37.08 %, 60.26 %, 78.80 %, respectively ($p<0.05$) with the addition of 5, 10, and 15 % fines. In the recycled beaten pulps, the addition of 5, 10, and 15 % fines caused an decrease in air permeance of 43.11 %, 59.89 %, and 85.33 %, respectively ($p<0,05$). For virgin pulps, the pulp freeness level had a more significant effect on air permeance than the content of fines, and vice versa for recycled pulp. For example, when the 15 % fines were added to Turkish Calabrian pine kraft pulp, hand sheet air permeance decreased from 5756 ml/min. to 857 ml/min. However, hand sheet air permeance was decreased with beating up to 28 ºSR from 5756 ml/min. to 126 ml/min. On the other hand, in the recycled pulp, hand sheet air permeance was decreased with beating up to 28 ºSR from 4925 ml/min. to 2032 ml/min, while hand sheet air permeance was decreased from 4925 ml/min. to 1133 ml/min with the addition of 15 % fines.

## 4 CONCLUSIONS
## 4. ZAKLJUČAK

Unless they are specifically removed, fines are present in any paper and influence its properties. In order to optimize the paper strength, it is important to control the fines content. In this study, the effect of fines on hand sheet properties was determined on different types of pulp (pine kraft pulp, aspen kraft pulp, and recycled pulp), as well as different pulp freeness levels (unbeaten pulp and 28 ºSR pulp). Based on the results, it can be concluded that secondary fines of recycled pulp have a significant impact on hand sheet properties. The effect of fines on hand sheet properties correlates strongly with the type of pulp, freeness level of pulp, and amount of fines addition. The effect of the addition of fines on hand sheet properties of unbeaten pulps was more prominent than of beaten pulps. On the other hand, effect of fines on hand sheet properties of the unbeaten and beaten pulps was dependent on the type of pulp. In the unbeaten and beaten pulp samples, the effect of fines addition was more pronounced in European aspen kraft pulp and recycled pulp than in Turkish Calabrian pine kraft pulp. The differences in hand sheet properties became more visible with the increasing fines content.

The highest paper strength increase rates were obtained from unbeaten pulps with 15 % fines. Paper strength of 15 % fines added unbeaten recycled pulp was higher than that of fines-free beaten recycled pulp. This demonstrates that it is possible to reduce the beating level by adding fines to recycled pulp. The results suggested that fines can be used as reinforcement for papermaking to improve the paper properties. In the papermaking process, fines can also be used as a potential control variable in order to obtain desirable paper properties. When the beating level and fines addition ratio are determined properly, refining energy requirements can be reduced, and paper strength can be improved.

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# Finite Element Modeling of Fiber Reinforced Polymer-Based Wood Composites Used in Furniture Construction Considering Semi-Rigid Connections

## Primjena metode konačnih elemenata za modeliranje drvno-plastičnih kompozita ojačanih vlaknima za uporabu u konstrukciji namještaja s polukrutim vezovima

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**ABSTRACT •** *In this study, control samples of pine (Pinus slyvestris L.), beech (Fagus orientalis L.) and oak (Quercus petreae L.) species were obtained by using fiber reinforced finger corner joints. Teknobont 200 epoxy and polyvinyl (PVAc) adhesives were used as glue. Bearing in mind the critical loads that may affect their use, experimental samples were tested under diagonal loads. Experimental samples were also analyzed by a computer program using the finite element method (FEM). Finally, experimental data were compared with the results of FEM. The comparisons clearly showed that experimental results and finite element solutions (SAP2000 V17) including semi-rigid connections are in good agreement. As a structural analysis program in furniture engineering designs, FEM can be preferred in terms of reliability and cost.*

***Keywords:*** *fiber reinforced polymer (FRP); furniture; glue; diagonal tension loading; finite element method (FEM)*

**SAŽETAK •** *U radu se prikazuju rezultati istraživanja uzoraka borovine (Pinus slyvestris L.), bukovine (Fagus orientalis L.) i hrastovine (Quercus petreae L.) spojenih kutnim zupčastim spojevima i ojačanih vlaknima. Kao ljepilo upotrijebljeno je epoksidno ljepilo Teknobont 200 i polivinilacetatno ljepilo (PVAc). Imajući na umu opterećenja koja se pojavljuju tijekom uporabe, eksperimentalni su uzorci ispitivani pri dijagonalnim opterećenjima. Uzorci su također analizirani računalnim programom primjenom metode konačnih elemenata (FEM). Eksperimentalni podatci i podatci dobiveni FEM analizom uspoređeni su te se jasno može vidjeti da se ti podatci za polukrute vezove podudaraju. Glede pouzdanosti i troškova, kao strukturnom programu analize u dizajniranju namještaja prednost se može dati FEM analizi.*

***Ključne riječi:*** *polimer ojačan vlaknima (FRP); namještaj; ljepilo; dijagonalno opterećenje; metoda konačnih elemenata (FEM)*

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## 1 INTRODUCTION
## 1. UVOD

In recent years, Fiber Reinforced Polymer (FRP) plates have been widely used because of their low density, due to their light weight, high resistance to corrosion and chemical effects, and easy application. Engineers and technical staff consistently work on concrete, steel, wood, stone, plastic, glass materials with the aim of obtaining various shapes and proportions of higher strength and more useful materials. In addition, new materials such as high strength glass, carbon, boron, aramide fiber have been developed recently.

In today's wooden structure design, the use of solid wood material, as one-piece in large-sized elements, is not feasible - both economically and technically. In addition, the use of single piece solid wood is limited in the production of load bearing elements. Complete removal of defects is not possible. This situation greatly affects the safety of the wooden structure. On the other hand, the use of solid single piece wood in the production of load bearing elements increases the rate of waste making it economically not viable. For this reason, in the design of wooden structures, it is possible to obtain the structural elements by joining the wood in the desired dimensions. However, the deformations caused by the service load in the wooden joints affect the material negatively. In order to eliminate this negative effect, studies on strengthening the joint regions should be carried out (Akgül, 2007).

In recent years, in the reinforcement of steel and reinforced concrete buildings, the number of applications with fiber reinforced plastics (FRP) in wood structures has been quite common. In wooden structural design, the size of the element depends on the proper joining details. In the performed studies, it has been determined that the joining areas of the designed wooden structures show high performance and that these regions are reinforced by using fiber reinforced plastics (glass reinforced composite plastic (GFRP), etc.) to increase the resistance to tension loads.

There are some reports in the literature on the effects of simulation of wooden materials in furniture construction (Gustafsson, 1995; Gustafsson, 1996; Gustafsson, 1997; Smardzewski, 1998; Smardzewski, 2002; Nicholls and Crisan, 2002; Guindos and Guaita, 2013; Tankut *et al*., 2014). Yorur (2012) reported that using FEM computer modeling enables faster, less costly, more optimized product development and examination of detailed product performance that cannot be observed experimentally. Nestorović *et al*. (2011) performed stiffness tests by using chair models and compared them with modeling analysis. The results of the study showed that the chair should not only be designed to achieve durability but also that the material should be designed properly and material properties should be changed. The literature on the use of finite element method in wooden construction is abundant, but there is not sufficient information on wood glass fiber reinforced material using FEM computer modeling.

In the skeletal system forming a wooden structure, the carrier elements are usually subjected to pressure, shrinkage and bending. Therefore, in this study, reinforcement of the wooden frame constructions, obtained by glass reinforced plastic (GRP) bars, was applied to the corner fasteners, which were subjected to pulling. The internal forces and deformations at the joints were determined by computer-aided structural analysis and then the theoretical and experimental deformations were compared.

## 2 MATERIALS AND METHODS
## 2. MATERIJALI I METODE

### 2.1 Wood
### 2.1. Drvo

Yellow pine, beech and oak wood used in the preparation of the test samples were obtained from the timber mills in Zonguldak region by random method. The wood samples used were kept in the climate room at a temperature of 20 ±2 ºC and relative humidity of 65 ±5 % until the air dried reached the required parameters.

### 2.2 Glue
### 2.2. Ljepilo

Epoxy glue: Teknobond 200 type epoxy, which is produced as a two-component bonding and assembly epoxy, was used for joining wooden surfaces and bonding GRP bars to wooden surfaces.

PVAc glue: It does not wear the cutting tools, it is odorless and non-flammable, cold applied, easy sliding and hardening.

### 2.3 Glass Fiber Reinforced Plastic (GFRP)
### 2.3. Plastika ojačana staklenim vlaknima (GFRP)

GFRP materials can be produced by various methods. The profile drawing method is used in CTP molding, especially in the construction sector, in the structure of profile type products, used as both the main material and complementary material. In addition to the box, pipe, I, T, L and U profiles produced by the profile drawing method, profiles with no fixed shape can be produced (Figure 1). In addition to the superior mechanical strength of GFRP material, its lightness, corrosion resistance, low density and good strength/density ratio, low thermal conductivity, lack of additional services such as maintenance and painting for many years, simple production with low labor force, and being easy to cut and machine, CTP profiles are advancing rapidly in the construction sector as an alternative to many materi-

**Figure 1** CTP profile samples produced by profile drawing method
**Slika 1.** CTP uzorci profila proizvedeni metodom crtanja profila

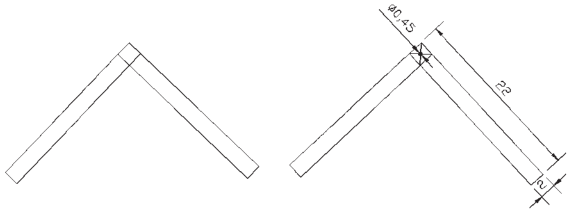


**Figure 2** Control and CTP as corner joining elements (cm)
**Slika 2.** Kontrolni i CTP kutni elementi (cm)

als (www.strongwell.com) due to the fact that they can be easily machined, complex geometry shapes can be easily produced, and they can be produced with different fiber layers and combinations to obtain different mechanical properties (www.strongwell.com).

### 2.4 Preparation of experiments
### 2.4. Priprema eksperimenta

Wood test specimens were prepared to be parallel to fiber directions from first class dried, cracked, knotless wood materials with dimensions of 20 mm × 46 mm and length of 220 mm. GFRP rods provided for the strengthening were cut to 5 cm each and placed in a form suitable for wood thickness. In this way, gear corner assemblies, which are especially used in frame constructions, were prepared. Adhesive was applied to the intersection surface of the prepared joints with a total of 160 g/m$^2$ for both types of glue. The diagonal tension loading was applied to the samples according to the principles set forth in ASTM-D 1037 (Figure 2). 8 samples were repeated in each group.

### 2.5 Test method
### 2.5. Metode ispitivanja

The diagonal drawing method, which represents the opening and closing of corner joints due to applied external forces, was determined as the test method (Figure 3). For the experiments, a universal test device was used in Bartin University Forest Faculty Laboratories. Static loading was carried out at a speed of 2 m/s. The maximum force values at the time of breaking or joining of the test specimens were recorded on the computer-programmable display connected to the test device.

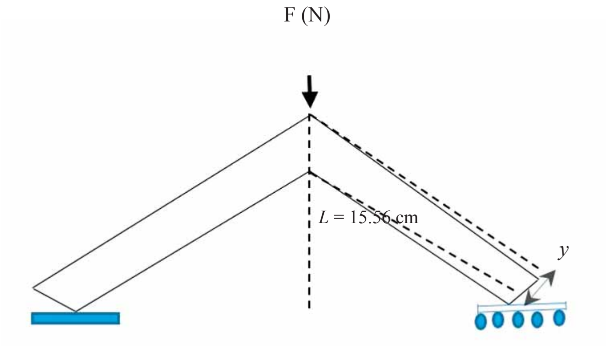


**Figure 3** Diagonal tension testing setup (cm)
**Slika 3.** Postavljanje dijagonalnog ispitivanja (cm)

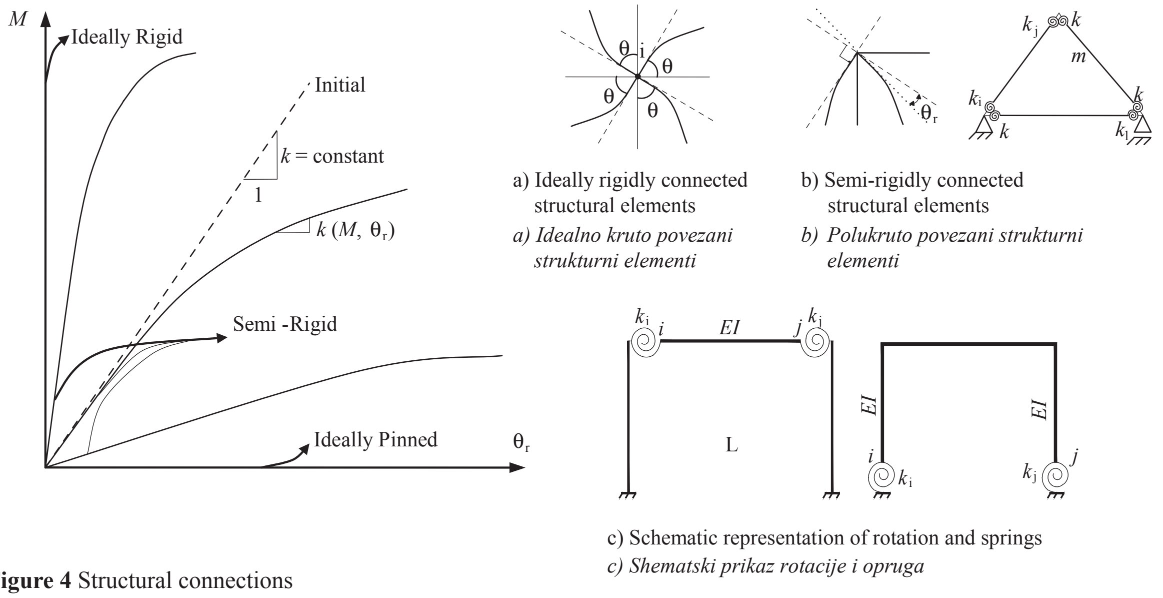
The difference of diagonal tension loading values in CTP samples with respect to the control samples was found by the formula in Eq. 1.

$$\%\textit{difference} = \frac{Cnt - CTP}{Cnt} \cdot 100 \qquad (1)$$

When there is a difference of diagonal tension loading values, *Cnt* represents the values of control samples and *CTP* the values of CTP samples.

### 2.6 Semi-rigid connection in finite element method
### 2.6. Polukruti vez u metodi konačnih elemenata

Structural elements and joints are designed based on some idealizations. The joints of idealized frame elements are assumed to be made by ideally rigid connections. However, another assumption is that structural members of truss systems have ideally pinned connection at joints. Actually, structural connections should be named according to their moment-rotation curves. These curves are usually derived by fitting suitable curves to the experimental data. Various types of M-$\theta_r$ models have been developed as described by Chen and Lui (1991). As seen from M-$\theta_r$ curves given in Figure 4, the



a) Ideally rigidly connected structural elements
*a) Idealno kruto povezani strukturni elementi*

b) Semi-rigidly connected structural elements
*b) Polukruto povezani strukturni elementi*

c) Schematic representation of rotation and springs
*c) Shematski prikaz rotacije i opruga*

**Figure 4** Structural connections
**Slika 4.** Strukturni vezovi

moment (*M*) is dependent on a function of relative rotation between structural members connected to the same joint. The finite element analyses are mostly performed assuming semi-rigid connections as rigid or pinned connections for simple calculation.

Connection flexibility is defined by various methods. To obtain an initial opinion on stiffness of rotational springs, the use of the modulus of elasticity (*E*), moment of inertia (*I*) and length (*L*) of related beam with constant cross-section is very effective and understandable. Stiffness matrix of a beam in local coordinates can be written using the attributes of this beam as follows (McGuire *et al*., 1999).

$$[k]=\begin{bmatrix} \frac{12EI}{L^3}\theta_1 & \frac{6EI}{L^2}\theta_2 & -\frac{12EI}{L^3}\theta_1 & \frac{6EI}{L^2}\theta_3 \\ \frac{6EI}{L^2}\theta_2 & \frac{4EI}{L}\theta_4 & -\frac{6EI}{L^2}\theta_2 & \frac{2EI}{L}\theta_5 \\ -\frac{12EI}{L^3}\theta_1 & -\frac{6EI}{L^2}\theta_2 & \frac{12EI}{L^3}\theta_1 & -\frac{6EI}{L^2}\theta_3 \\ \frac{6EI}{L^2}\theta_3 & \frac{2EI}{L}\theta_5 & -\frac{6EI}{L^2}\theta_3 & \frac{4EI}{L}\theta_6 \end{bmatrix} \tag{2}$$

Where $\theta_{1\text{-}6}$ are the coefficients given as follows:

$$\theta_1=\frac{\alpha_i+\alpha_j+\alpha_i\alpha_j}{4(3+\alpha_j)+\alpha_i(4+\alpha_j)} \tag{3.a}$$

$$\theta_2=\frac{\alpha_i(2+\alpha_j)}{4(3+\alpha_j)+\alpha_i(4+\alpha_j)} \tag{3.b}$$

$$\theta_3=\frac{\alpha_j(2+\alpha_i)}{4(3+\alpha_j)+\alpha_i(4+\alpha_j)} \tag{3.c}$$

$$\theta_4=\frac{\alpha_i(3+\alpha_j)}{4(3+\alpha_i)+\alpha_j(4+\alpha_i)} \tag{3.d}$$

$$\theta_5=\frac{\alpha_i\alpha_j}{4(3+\alpha_i)+\alpha_j(4+\alpha_i)} \tag{3.e}$$

$$\theta_6=\frac{\alpha_j(3+\alpha_i)}{4(3+\alpha_i)+\alpha_j(4+\alpha_i)} \tag{3.f}$$

Here, $\alpha_i$ and $\alpha_j$ are the stiffness indexes and can be used to obtain rotational spring stiffness as follows:

$$k_i=\alpha_i\frac{EI}{L} \tag{4.a}$$

$$k_j=\alpha_j\frac{EI}{L} \tag{4.b}$$

Where, $k_i$ and $k_j$ are the rotational spring stiffness at *i* and *j* ends of the beam, respectively, and those change in 0-∞ range.

Semi-rigid connection may also be identified by connection percentage. Then, the parameters of $\theta_i$ can be written as follows (Chen and Lui, 1991; Kartal, 2004; Filho *et al*., 2004):

$$\theta_1=\frac{r_i+r_j+r_{ij}}{3} \tag{5.a}$$

$$\theta_2=\frac{2r_i+r_{ij}}{3} \tag{5.b}$$

$$\theta_3=\frac{2r_j+r_{ij}}{3} \tag{5.c}$$

$$\theta_4=r_i \tag{5.d}$$

$$\theta_5=r_{ij} \tag{5.e}$$

$$\theta_6=r_j \tag{5.f}$$

Where, $r_i$, $r_j$ and $r_{ij}$ are the correction factors obtained as follows:

$$r_i=\frac{3\cdot\nu_i}{4-\nu_i\cdot\nu_j} \tag{6.a}$$

$$r_j=\frac{3\cdot\nu_j}{4-\nu_i\cdot\nu_j} \tag{6.b}$$

$$r_{ij}=\frac{3\cdot\nu_i\cdot\nu_j}{4-\nu_i\cdot\nu_j} \tag{6.c}$$

Here, $\nu_i$ and $\nu_j$ are the fixity factors and represent the semi-rigid connection as percentage. If the Eq. 3 and 5 are equalized, a set of equations, which provides a direct relation with initial spring stiffness and connection percentage, is achieved as presented in Eq. 7 (Monforton and Wu, 1963; Sekulovic and Salatic, 2001),

$$k_{i,j}=\frac{3\cdot EI\cdot\nu_{i,j}}{(1-\nu_{i,j})\cdot L} \tag{7}$$

Where $\nu_{i,j}$ is the fixity factor, which represents the connection percentage.

After the stiffness matrix [K] and force vector {F} of the system are formed, the displacement vector {U} is obtained from Eq. 7.

$$\{F\}=[K]\{U\} \tag{8}$$

Then the internal forces and moments occurring in the structure, including semi-rigid connections, may be easily acquired.

## 3 RESULTS AND DISCUSSION
## 3. REZULTATI I RASPRAVA

The average maximum fracture load in diagonal tension loading obtained from the glue combinations of wood species used in the experiments are given in Figure 5.

As shown in Figure 5, the diagonal shrinkage value of oak species combined with PVAc glue, with respect to the control samples, shows a decrease of about 3 % in CTPs. When combined with epoxy glue, the diagonal tension loading of oak species increases by about 8 % in CTPs. With respect to the control samples, the diagonal shrinkage value of yellow pine species combined with PVAc glue decreases by approximately 9 %. When the connection with epoxy glue is provided, the maximum fracture load in diagonal tension loading of yellow pine species shows a decrease of about 32 % in CTPs. With respect to the control samples, the average maximum fracture load in diagonal tension loading of beech species combined with

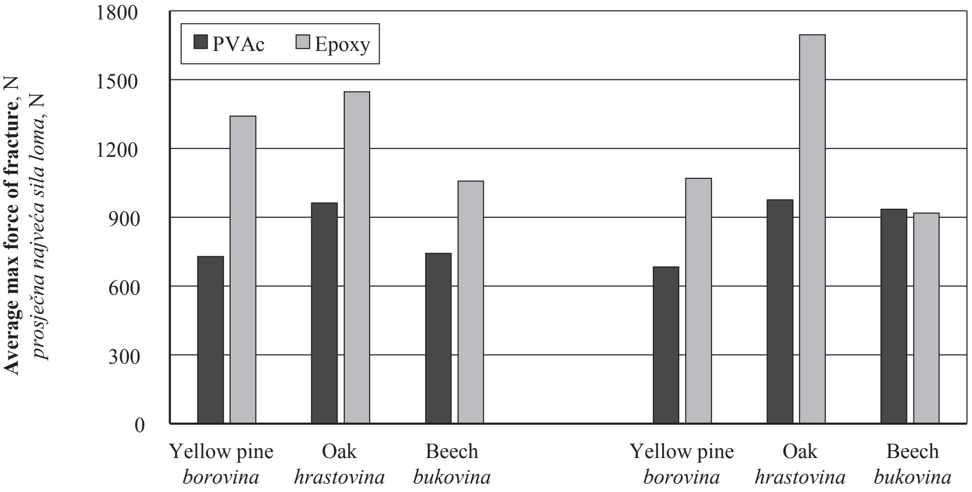


**Figure 5** Average max force of fracture in diagonal tension loading
**Slika 5.** Prosječna najveća sila loma pri dijagonalnom opterećenju

PVAc glue increases by about 30 % in the GFRP samples. When combined with epoxy glue, the average of maximum fracture load in diagonal tension loading of beech species is reduced by about 11 % in CTPs. As a result of the strengthening process, a good result was obtained for oak species combined with epoxy with the combination of beech and PVAc.

The deformations were obtained as a result of finite element analysis carried out using the SAP2000 program under certain load for the wooden frame system. The comparisons of deformation for PVAc and epoxy groups are shown in Figure 6 and Figure 7.

As seen in Figure 6, the results obtained from the experiments of PVAc with respect to the results of

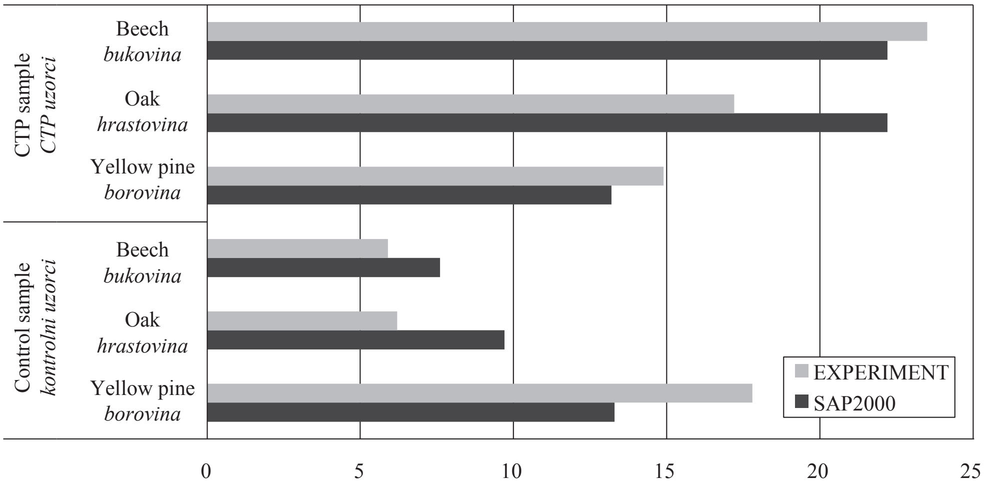


**Figure 6** Comparison of PVAc group values obtained by SAP2000 program with experimental results
**Slika 6.** Usporedba vrijednosti PVAc grupe dobivenih uz pomoć programa SAP2000 s eksperimentalnim rezultatima

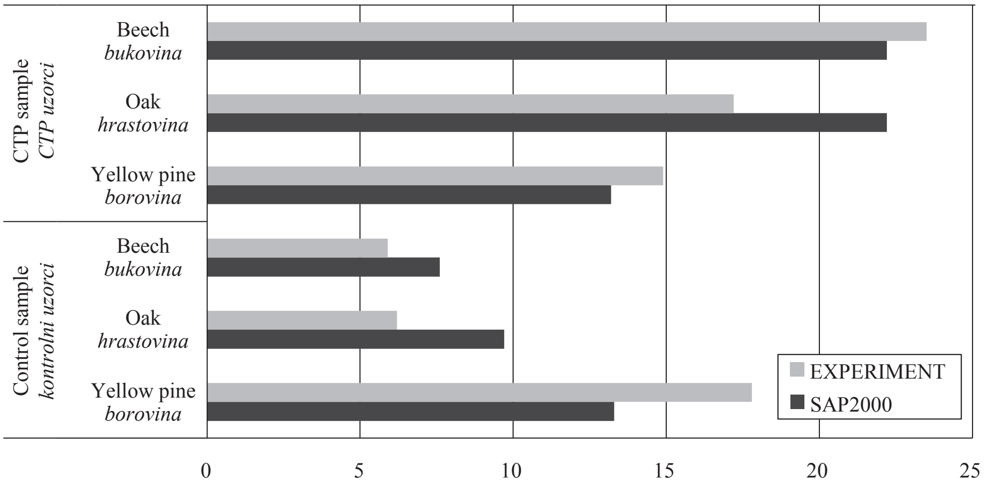


**Figure 7** Comparison of epoxy group values obtained by SAP2000 program with experimental results
**Slika 7.** Usporedba vrijednosti *epoxy* grupe dobivenih uz pomoć programa SAP2000 s eksperimentalnim rezultatima

**Table 1** Material properties used in SAP2000
**Tablica 1.** Svojstva materijala korištenih u programu SAP2000

| **Wood type**<br>*Vrsta drva* | **Density, g/cm$^3$**<br>*Gustoća,* g/cm$^3$ | **Modulus of elasticity, N/mm$^2$**<br>*Modul elastičnosti,* N/mm$^2$ | **Length of material, mm**<br>*Dužina uzorka,* mm | **Poisson ratio**<br>*Poissonov omjer* |
|---|---|---|---|---|
| Yellow Pine / *žuti bor* | 0.51 | 11760 | 220 | 0.30 |
| Oak / *hrast* | 0.71 | 12500 | 220 | |
| Beech / *bukva* | 0.65 | 12250 | 220 | |

SAP2000 show an agreement of approximately 80-90 %. It was observed that the control samples had less deformation than CTP samples.

As seen in Figure 7, the results obtained from the epoxy group experiments are close to 70-80 % with respect to the results of SAP2000. The control samples showed less deformation than the GFRP samples. At the joining point of CTP bars, the deformation values were found to be higher than those of the control samples and the strength was low.

Static analysis of the load bearing systems consisting of wooden bar elements made of yellow pine, beech and oak were performed. In the analyses, the load effects of the wooden structural elements and the load effects applied in experimental studies were taken into consideration. Table 1 presents the properties of the materials used for wooden structural elements. In the finite element analyses made by SAP2000 program, with the application of FEM, semi-rigidity was used. The geometry of the experimental models used in this study is constant and the finite element model representing these models is given in Figure 8.

In the FEM, yellow pine wooden frame system is considered as control group. In this case, separate combination percentages of PVAc and epoxy glues were determined for yellow pine group. Then, using these percentages, the rotational spring stiffness of the rod ends of the other wood groups were calculated and finite element analysis was performed. As a result of the analysis, comparison was made with the vertical deformation values obtained from the experimental results as shown in Table 2. The same solution algorithm was also performed for GFRP reinforced timber bar systems.

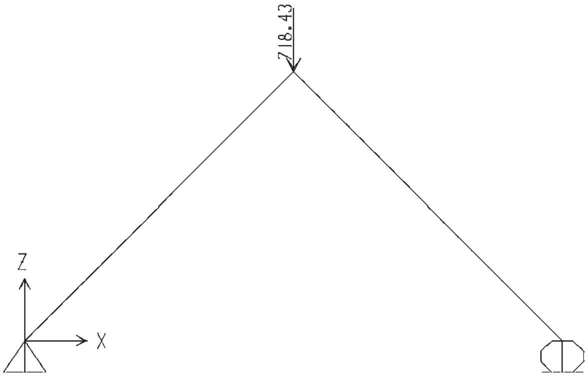


**Figure 8** Finite element model of beech wood frame system and loading case (N)
**Slika 8.** Model konačnih elemenata okvira od bukovine tijekom opterećenja (N)

The finite element analysis and the deformation values obtained from experimental studies are seen in good agreement - about 80-90 %.

## 4 CONCLUSIONS
## 4. ZAKLJUČAK

According to the experimental results, a slightly higher diagonal tension loading was obtained from oak combined with epoxy glue and beech combined with PVAc as a result of the strengthening process. When the circle type is selected in the CTP rod corner as the joining element, it is seen that it does not give a good result in the diagonal tension loading.

In this study, wood frame construction was modeled with the SAP2000 finite element program and the

**Table 2** Comparison of experimental results with SAP2000 program
**Tablica 2.** Usporedba eksperimentalnih rezultata s rezultatima dobivenim programom SAP2000

| **Groups**<br>*Grupe* | **Wood type**<br>*Vrsta drva* | **Glue type**<br>*Vrsta ljepila* | **Vertical deformation, mm**<br>*Vertikalna deformacija,* mm | |
|---|---|---|---|---|
| | | | **Experimental results**<br>*Eksperimentalni rezultati* | **SAP2000 Results**<br>*Rezultati programa SAP2000* |
| **Control samples**<br>*Kontrolni uzorci* | Yellow pine / *borovina* | PVAc | 14.49 | 14.42 |
| | | Epoxy | 8.15 | 8.48 |
| | Oak / *hrastovina* | PVAc | 18.03 | 17.77 |
| | | Epoxy | 8.5 | 9.96 |
| | Beech / *bukovina* | PVAc | 13.07 | 13.19 |
| | | Epoxy | 5.5 | 7.3 |
| **CTP samples**<br>*CTP uzorci* | Yellow pine / *borovina* | PVAc | 17.37 | 17.06 |
| | | Epoxy | 17.62 | 17.79 |
| | Oak / *hrastovina* | PVAc | 24.83 | 22.29 |
| | | Epoxy | 10.82 | 33.45 |
| | Beech / *bukovina* | PVAc | 22.62 | 22.17 |
| | | Epoxy | 22.2 | 14.66 |

obtained analysis results were compared with the results obtained from the experimental study. In the finite element solutions, the rotational spring stiffness of the wooden elements at the connection points is considered as a variable parameter. Based on the above solutions, the results of the SAP2000 analysis showed an agreement of 80-90 % in combination with PVAc glue and 70-80 % in combination with epoxy glue. This case shows that approximate results can be obtained by the boundary conditions of the computer model of experimental mechanism. The boundary conditions are easily applied in the computer program and the restrictions can be met when creating the experimental setup. Inevitably, there can be some mismatch between the experiment and the model in terms of the bearing conditions, initial conditions and approximations. Therefore, this situation leads to a discrepancy in the results. In terms of the bending moment bearing capacity, it may be assumed that the corner connection point should be further tested with the L-type or T-type bars instead of the circular bars. Furthermore, by determining the ratio of partial fixity for each material, the differences between experimental and numerical results can be clearly decreased for further expanded studies.

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# LABORATORY FOR HYDROTHERMAL PROCESSING OF WOOD AND WOODEN MATERIALS

Testing of hydrothermal processes of wood and wooden materials

Thermography measurement in hydrothermal processes

Standard and nonstandard determination of moisture content in wood

Determination of climate and microclimate conditions in air drying and storage of wood, organization of lumber storage

Project and development of conventional and unconventional drying systems

Steaming chamber projects

Establishing and modification of kiln drying schedules

Consulting in selection of kiln drying technology

Introduction of drying quality standards

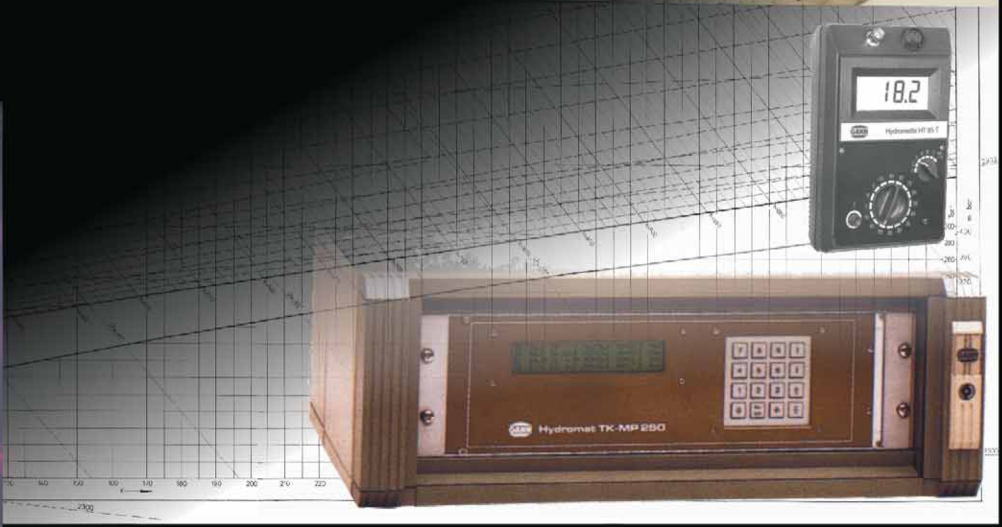
Determination of wood bending parameters

Detection and reducing of hydrothermal processes wood defects

Reducing of kiln drying time

Drying costs calculation

Kiln dryer capacity calculation



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# Improving Weathering Performance of Wood by Borates Impregnation and Liquid Glass Coating

## Poboljšanje otpornosti drva na vremenske utjecaje impregnacijom boratima i premazom od tekućeg stakla

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**ABSTRACT •** *Weathering performance of impregnated and coated wood products is an important issue that influences their appearance as well as their service life after outdoor or indoor exposure. A novel procedure to improve the weathering performance of Scots pine wood (Pinus sylvestris L.) is proposed in this study. Wood samples were impregnated with ammonium tetrafluoroborate (ATFB), ammonium pentaborate (APB) and boric acid (BA), and afterward coated with a layer of liquid glass (LG). Surface hardness, color changes, and surface roughness of wood samples were investigated after 250 h and 500 h of exposure to artificial weathering. The results revealed that, except for untreated (control), all other treatment groups caused an increase in surface hardness of Scots pine after weathering. A decrease in the CIE L* value corresponds to the darkening of samples after weathering. Borates-impregnated and LG-coated Scots pine samples possessed the highest color stability. However, all treatment combinations resulted in reddish and yellowish tones after weathering. Exposure to weathering increased surface roughness of wood samples. The surface roughness of the untreated (control) group was higher than that of impregnated and coated samples.*

***Keywords:*** *weathering; boron compounds; liquid glass; wood coating; surface characteristics; building material*

**SAŽETAK •** *Otpornost impregniranoga i premazanog drva pri izlaganju vremenskim utjecajima velik je problem koji utječe na izgled i vijek trajanja drva nakon izlaganja u eksterijeru ili interijeru. U ovom je istraživanju predložen novi postupak poboljšanja otpornosti borovine (Pinus sylvestris L.) izložene vremenskim utjecajima. Uzorci drva impregnirani su amonijevim tetrafluoroboratom (ATFB), amonijevim pentaboratom (APB) i bornom kiselinom (BA), nakon čega su premazani tekućim staklom (LG). Ispitivana je tvrdoća površine, promjena boje i hrapavost površine uzoraka drva nakon 250 i 500 sati umjetnog izlaganja vremenskim utjecajima. Rezultati su pokazali da se na svim površinski obrađenim uzorcima borovine povećala tvrdoća njezine površine nakon izlaganja vremenskim utjecajima. Smanjenje vrijednosti CIE L* upućuje na tamnjenje uzoraka nakon izlaganja vremenskim*

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*utjecajima. Najbolju stabilnost boje pokazali su uzorci borovine impregnirani boratima i premazani tekućim staklom. Međutim, sve kombinacije površinske obrade rezultirale su povećanjem crvenoga i žutog tona nakon izlaganja uzoraka vremenskim utjecajima. Izlaganjem vremenskim utjecajima povećala se hrapavost površine uzoraka drva. No hrapavost površine kontrolnih uzoraka (površinski neobrađenih) bila je veća nego hrapavost površine impregniranih i premazanih uzoraka.*

***Ključne riječi:*** *izlaganje vremenskim utjecajima; spojevi bora; tekuće staklo; premaz za drvo; svojstva površine; građevni materijal*

## 1 INTRODUCTION
## 1. UVOD

Wood is available throughout the world in both natural and plantation forests, and it is used in a wide range of consumer products, particularly in the interior and exterior building and construction applications (Woodard and Milner, 2016). It was proven that the quality and aesthetic appeal of the value-added wood-based products can be competitive to other non-renewable materials available on the market, determining its economic success (Fell, 2002). Certain coatings, such as paints and varnishes, are commonly used to protect wood against weathering in an exterior environment and to enhance its natural appearance (Cristea *et al.,* 2010). The aesthetic value of wood can quickly be lost if it is not protected from the weather. This can be solved by applying transparent protective finishes or opaque coatings. Clear finishes highlight the natural color and texture of wood, but penetration of solar radiation can degrade the underlying surface and cause the peeling of the finish (Pandey and Pitman, 2002). Special attention should, therefore, be directed to preserve the color of wood-based products because this property adds particular value to the product. Various chemical reactions that cause color and appearance changes are induced by diverse abiotic factors (David *et al*., 2000). Weathering is one of the major abiotic factors affecting the original color and natural appearance of wood. Ultraviolet (UV) radiation causes the wood color to turn gray, and with constant exposure, the fibers of the wood surface weaken due to depolymerization of lignin and carbohydrates in the cell wall. When wood is exposed to sunlight, lignin absorbs UV light and is responsible for color changes in these wood components. Fungal growth, warping, checking, and splitting can also occur on the wooden surface after a specific exposure time (Mohebby and Saei, 2015; Ghosh *et al.,* 2009).

The color change of heat-treated wood species (ash, iroko, Scots pine, and spruce) was investigated during natural weathering. It was found that there was apparent color fading of wood surfaces after exposure (Tomak *et al*., 2014). The weathering process was primarily linked to the sunlight irradiation and its rate was increased by higher air temperature, moisture, and other pollutants in the outdoor environment (Pandey, 2005). Accordingly, ultraviolet (UV) radiation and moisture were identified as the main causes of deterioration and discoloration of wood surfaces in weather conditions (Huang *et al*., 2012; Berdahl, 2008). Coating of the wood surface may be considered as a solution to reduce the effects of weather conditions on material deterioration and to preserve the aesthetic properties of the material exposed outdoor in different cardinal directions or varying design details (Herrera *et al.,* 2018). Weathering is defined as the slow decomposition of materials exposed to weather factors (Sandak *et al*., 2015). Weathering effects can be observed depending on the type of wood or process. Although natural weathering is usually a relatively slow process, artificial weathering plays an important role in evaluating coatings by obtaining performance assessments in much shorter times than natural weathering. While artificial weathering methods do not fully represent actual material changes in certain weather conditions, they are generally considered to be a very useful tool for predicting surface resistance and future product performance (Evans *et al.,* 2005; Jankowska and Kozakiewicz, 2014). Accordingly, ultraviolet (UV) radiation and moisture were identified as the main causes of deterioration and discoloration of wood surfaces in weather conditions (Huang *et al*., 2012; Berdahl, 2008).

In the wood preservation industry, traditional wood preservatives are used to prevent the degradation of wood by biological organisms. However, many common wood preservatives, such as copper chromium arsenate (CCA), are prohibited due to their harmful environmental effects. In recent years, research has produced a new generation of improved wood preservatives to extend the service life of wood material. Natural extracts are among the most important environmentally friendly wood preservatives, and many studies have reported that natural plant extracts can protect wood against biological organisms (Salem *et al.,* 2016).

Boron compounds have been recognized and accepted as wood preservatives against insects and fungi (Thevenon *et al.,* 1997). Boron compounds are relatively cost-effective. Despite their many advantages, boron compounds cannot provide complete protection for outdoor applications due to their low leaching performance (Kartal and Green, 2002; Kartal and Imamura, 2004).

Liquid glass (LG) is a two-component product used for waterproofing of surfaces requiring high chemical and physical resistance. It possesses excellent water resistance and can be produced as a transparent or color product that is resistant to atmospheric conditions. LG is not affected by UV light radiation and outdoor weather conditions (http://www.isonem.com, 2019).

The durability of the material and its performance against weathering play an important role in material selection in the building and furniture industry. Customers expect environmentally friendly, sustainable, aesthetic, and durable wood-based materials for interior and exterior spaces.  Hardness, color, and roughness of the wood surface are, therefore, important parameters in the construction and building sector. Architects demand high-performance wooden materials for their design. Boron compounds are one of the most environmentally friendly, nontoxic for human health, and advanced impregnation materials. This study aimed to improve the weathering performance of borates-impregnated and LG coated Scots pine wood according to market expectations.

## 2 MATERIALS AND METHODS
## 2. MATERIJALI I METODE

### 2.1 Materials
### 2.1. Materijali

### 2.1.1 Experimental samples
### 2.1.1. Eksperimentalni uzorci

Straight-grained and defect-free sapwood samples of Scots pine (*Pinus sylvestris* L.) wood were prepared according to ASTM D 358-98 standard (1998). The approximate initial moisture content of the samples was 12 % and the dimensions of samples used in the experiment were 6 mm (radial) × 75 mm (tangential) × 150 mm (longitudinal). Each sample was sanded with P180 grit sandpaper to obtain a smooth surface. Then wood samples were oven dried at 50 ± 5 °C until constant weight. Ten replicate samples ($n$ = 10) were prepared for each surface finishing variation.

### 2.1.2 Impregnation procedure
### 2.1.2. Postupak impregnacije

The test samples were impregnated with 4 % aqueous solution of ammonium tetrafluoroborate (ATFB), ammonium pentaborate (APB), and boric acid (BA) according to ASTM D1413-07e1 (2007). A vacuum desiccator was used for the impregnation process. It was connected to a vacuum pump through a vacuum trap. The vacuum was carried out at 760 mm Hg for 30 minutes before supplying the solution into the chamber, followed by a further 30 minutes at 760 mm Hg diffusion time under vacuum.

Retention $R$ values of chemicals were calculated using Eq. 1:

$$\text{Retention} = \frac{G \cdot C}{V} \cdot 10^3 \text{ (kg / m}^3\text{)} \qquad (1)$$

Where: $G = (T_2 - T_1)$ – mass of impregnating solution absorbed by wood sample (g)
$T_1$ – mass of wood sample before impregnation (g)
$T_2$ – mass of wood sample after impregnation (g)
$C$ - concentration of impregnating solution (%)
$V$ - volume of wood specimen (cm$^3$)

### 2.1.3 Liquid glass
### 2.1.3. Tekuće staklo

Liquid glass (LG), used for experimental samples coating, was provided by Isonem Liquid Glass Paint and Insulation Technologies in Izmir, Turkey, a construction industry trade joint-stock company.

LG was applied to the surfaces of test samples according to the manufacturers’ instructions. The viscosity of the LG was 18 s via using DIN 4 cup at 20 ± 2 °C as recommended by the coating manufacturer. A and B component ratios were determined 7/1, respectively, according to the manufacturer’s suggestions.

The LG was applied to all surfaces and sides of the wood samples with a spray gun according to ASTM D3023-98 standard (2013). The filler was not used to avoid potential interference with the surface characteristics of wood. Instead of using the filler, LG was applied twice. Spreading rate and time were 200 g/m$^2$ and 48 hours, respectively. After the application of the top coating of LG to the surfaces, wood specimens were conditioned at 20 °C and 65 % relative humidity for 3 weeks.

### 2.2 Methods
### 2.2. Metode

Non-toxicity, fire resistance, aesthetics, and durability are among the desired material characteristics. Therefore, borates, which have high fire resistance and are not harmful to human health, were selected to impregnate Scots pine wood in this research.

In this study, impregnated and coated Scots pine wood was exposed to artificial weathering. Selected surface properties, such as hardness, color and roughness were investigated along the experiment to understand the changes of some physical and aesthetics characteristics. The arithmetic mean and standard deviation were calculated as a measure of repeatability and fit of experimental results.

### 2.2.1 Artificial weathering
### 2.2.1. Umjetno izlaganje vremenskim utjecajima

The artificial weathering test was performed according to ASTM G154-06 standard (2016) in a QUV weathering tester with eight UVA 340 lamps. Wood samples were exposed to artificial weathering for 250 h and 500 h, corresponding to light irradiation of 8 h, followed by a de-ionized water condensation for 4 h cycles, respectively. The chamber temperatures during the light irradiation and condensation periods were 60 °C and 50 °C, respectively. The average irradiance was 0.89 Wm$^2$/nm$^1$ at 340 nm of light wavelength.

### 2.2.2 Surface hardness test
### 2.2.2. Mjerenje tvrdoće površine

The surface hardness of Scots pine wood samples was measured with the König hardness according to ASTM D 4366–14 (2014). The specimen was positioned on a panel table and a pendulum was placed on the panel surface. The pendulum was deflected for 6° and released, triggering the start of the timer. The time needed to decrease the pendulum amplitude from 6° to 3° corresponded to the König hardness estimate.

### 2.2.3 Color test
### 2.2.3. Mjerenje boje

The color test parameters $L^*$, $a^*$, and $b^*$ were determined according to the *CIE* $L^*a^*b^*$ method. The $L^*$ color coordinate represents the lightness, whereas $a^*$ and $b^*$ are related to the chromaticity. The $L^*$ value can vary from 100 (white) to 0 (black). The value of $a^*$ represents red (positive) or green (negative) tones. Correspondingly, the positive $b^*$ parameter corresponds to yellow, whereas negative $b^*$ represents blue. (Zhang, 2003).

The color of test samples was measured by the X-Rite colorimeter (SP Series Spectrophotometer, X-ride Pantone, USA) before and after both artificial weathering periods. The total color change (ΔE*) was determined for each wood material according to ASTM D1536-58T (1964). The observer angle was 10° and the standard illuminant was D65.

The color changes were calculated according to Eqs. 2 to 5.

$$\Delta a^* = a_f^* - a_i^* \tag{2}$$

$$\Delta b^* = b_f^* - b_i^* \tag{3}$$

$$\Delta L^* = L_f^* - L_i^* \tag{4}$$

$$\Delta E^* = \sqrt{(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2} \tag{5}$$

Where $\Delta a^*$, $\Delta b^*$ and $\Delta L^*$ represent the differences between the color coordinates before and after artificial weathering.

### 2.2.4 Surface roughness test
### 2.2.4. Mjerenje hrapavosti površine

The surface roughness of the wood samples was measured by the Mitutoyo Surftest SJ-301 (Mitutoyo Corporation. Tokyo. Japan) according to DIN 4768 (1990). The surface roughness profile was scrutinized by means of the stylus with a diamond tip of 5 μm radius and 90° conical angle. The feed speed of the stylus was 0.5 mm/s[1] along 8 mm sampling length (Zhong *et al*., 2013). Three parameters, which are typically used for wood roughness quantification, were computed, including mean arithmetic deviation of the profile (*Ra*), mean peak-to-valley height (*Rz*), and root mean square (*Rq*) (Hiziroglu and Graham, 1998; Zhong *et al*., 2013). The cut-off of the filter was 2.5 mm.

## 3 RESULTS AND DISCUSSION
## 3. REZULTATI I RASPRAVA

### 3.1 Retentions of wood samples
### 3.1. Retencija uzoraka drva

The retention results of Scots pine wood impregnated with 4 % impregnation solution of chemicals are shown in Table 1. The highest amount of retention was noticed in samples impregnated with BA, while the lowest amount of retention was obtained in the experimental samples impregnated with APB.

### 3.2 Surface hardness
### 3.2. Hrapavost površine

Surface hardness values of the borates-impregnated and LG-coated Scots pine wood samples before and after the artificial weathering process are summarized in Table 2.

Borates impregnation and LG coating increased the hardness of Scots pine surface before artificial weathering. The hardness of untreated (control) wood was 29.3 and increased to 49.4 and 64.4, respectively, after impregnation and LG coating. This might be due to the hardness properties and composition of borates and LG. It was also observed that the surface hardness of the untreated Scots pine decreased after artificial weathering.

The results of the surface hardness of wood materials after weathering obtained in this experiment are in line with other scientific studies. Softening of the untreated Scots pine wood surface and decrease of its hardness after weathering was reported by several re-

**Table 1** Retentions of wood samples
**Tablica 1.** Retencija uzoraka drva

| **Impregnation solution** *Otopina za impregnaciju* | **Concentration** *Koncentacija* % | **Retention** *Retencija* kg/m³ | **Standard deviation** *Standardna devijacija* |
|---|---|---|---|
| ATFB | 4 | 24.82 | 5.6 |
| APB | 4 | 22.94 | 3.9 |
| BA | 4 | 25.77 | 4.3 |

**Table 2** Surface hardness values of samples before and after artificial weathering
**Tablica 2.** Vrijednosti hrapavosti površine uzoraka prije i nakon umjetnog izlaganja vremenskim utjecajima

| **Impregnation and coating types** *Vrste impregnacije i premaza* | **Before artificial weathering** *Prije umjetnog izlaganja vremenskim utjecajima* | | **After 250 h artificial weathering** *Nakon 250 h umjetnog izlaganja vremenskim utjecajima* | | **After 500 h artificial weathering** *Nakon 500 h umjetnog izlaganja vremenskim utjecajima* | | **Change after 250 hours** *Promjena nakon 250 h* | **Change after 500 hours** *Promjena nakon 500 h* |
|---|---|---|---|---|---|---|---|---|
| | Mean | *SD* | Mean | *SD* | Mean | *SD* | % | % |
| Control / *kontrolni uzorci* | 29.33 | 2.80 | 23.83 | 3.43 | 23.67 | 1.86 | -18.75 | -19.32 |
| ATFB + Liquid glass<br>*ATFB + tekuće staklo* | 49.60 | 3.36 | 62.40 | 4.34 | 70.20 | 5.72 | 25.80 | 41.53 |
| APB + Liquid glass<br>*APB + tekuće staklo* | 49.40 | 4.16 | 71.80 | 2.49 | 73.00 | 6.67 | 45.34 | 47.77 |
| BA + Liquid glass<br>*BA + tekuće staklo* | 64.40 | 1.95 | 55.80 | 4.49 | 56.00 | 4.64 | 13.35 | 13.04 |
| Liquid glass<br>*tekuće staklo* | 53.60 | 4.04 | 78.80 | 6.98 | 65.80 | 4.82 | 47.01 | 22.76 |

*SD* – Standard deviation / *standardna devijacija* searchers (Ustun *et al*., 2016; Turkoglu *et al*., 2015; Baysal, 2012; Yalinkilic *et al*., 1999). However, the opposite trend was observed for impregnated and LG coated samples investigated in this test, where hardening of wood surface occurred after 250 h and 500 h artificial weathering. The highest surface hardness increase was noticed for LG-coated Scots pine after 250 h of exposure. However, it decreased after continuous weathering. Surface hardness increased after 500 h artificial weathering for APB and ATFB-impregnated and LG-coated samples. After 500 h artificial weathering, the highest hardness was noticed for APB-impregnated and LG-coated Scots pine.

Cakicier *et al*. (2011) found that the surface hardness of wood samples coated with water-based varnish increased after artificial weathering for 432 h. They attributed this increase to some modifications in molecular cohesion induced by UV rays, which has a great effect on varnish layer hardness. Polymerization reactions occur and crosslinks are formed between the varnish molecules as the large molecules are cured, which increases the hardness of the wood coating system. De Meijer (2002) claimed that an increase in the temperature of thermal weathering could increase molecular cohesion among the resin molecules of the varnish layers. Baysal (2008) found that synthetic varnish (SV) and polyurethane varnish (PV) coating together with copper chrome boron (CCB) impregnation hardened wood surfaces after 3 months of outdoor natural weathering. In another study, Baysal (2011) reported that the increase in wood surface hardness of CCB-impregnated and synthetic varnish-coated samples was higher than of those treated with CCB-impregnated and coated with polyurethane varnish. The analysis was performed after artificial weathering exposure for 500 h. Baysal *et al.* (2014) investigated surface hardness of Scots pine pre-impregnated with copper-based chemicals before varnish coating after 500 h artificial weathering. They found that all impregnated and coated wood increased its surface hardness after 500 h artificial weathering. The experimental results reported here are therefore in good agreement with these researchers' findings.

### 3.3 Color change
### 3.3. Promjena boje

Table 3 shows $L^*$, $a^*$, and $b^*$ values of untreated (control) and impregnated and LG-coated Scots pine samples before and after artificial weathering. The table presents changes for each color coordinate ($\Delta L^*$, $\Delta a^*$, and $\Delta b^*$) as well as the total color change ($\Delta E^*$) for wood samples after 250 and 500 h artificial weathering. Before artificial weathering, all treatment combinations showed a decrease of $L^*$ values (62.97 to 68.63) when compared to the untreated (control) samples (75.69). Before artificial weathering, lightness $L^*$ of LG-coated (not-impregnated) Scots pine was higher than that of borates-impregnated and LG-coated samples. It indicates a more profound darkening of wood after the borates treatment. These results are in good agreement with those of Baysal (2012), Ustun *et al.* (2016), and Simsek and Baysal (2012), who investigated the effects of some impregnation materials on color changes of wood surface.

Results of the test revealed that, while $a^*$ value of control was higher than that of other treatment groups, $b^*$ values of borates-impregnated and LG-coated Scots pine were higher than those of the control group. *CIE* $a^*$ and $b^*$ values of un-treated Scots pine (control) were 5.9 and 54.6 before artificial weathering. While $a^*$ values changed from 6.6 to 9.4, $b^*$ values changed from 48.0 to 53.0 for borates-impregnated and LG-coated Scots pine.

A negative lightness change ($\Delta L^*$) occurred after 250 and 500 h of artificial weathering for un-treated (control) and all borates-impregnated and LG-coated Scots pine samples. $\Delta L^*$ is the most sensitive parameter describing color changes of wood surface. Therefore, the wood surface became rougher and darker after weathering. The darkening of Scots pine due to weathering is related to the photodegradation and leaching of lignin and other non-cellulosic polysaccharides (Sonmez *et al*., 2011; Petric *et al*., 2004; Hon and Chang, 1985). While positive values of $\Delta a^*$ indicate a tendency of the Scots pine wood surface to become reddish, positive values of $\Delta b^*$ indicate that the Scots pine wood surface tends to turn yellow. After 250 and 500 h of weathering, positive $\Delta a^*$ and $\Delta b^*$ indicate that the investigated wood surfaces maintained a reddish and yellowish tone after weathering.

Total color change $\Delta E^*$ of untreated (control) was between 20.6 and 22.7 after 250 and 500 h of weathering, respectively. The reduction of $\Delta E^*$ in borates-impregnated and LG-coated woods suggests a positive contribution of finishing and improved color stability. The best stability after 500 h of artificial weathering was noticed in APB-impregnated and LG-coated Scots pine. Borates impregnation before LG-coating stabilized color changes due to weathering. It might be due to the photo stabilization of wood via borate treatments, which could be explained by retardation of the carbonyl group formation and reduced delignification due to weathering (Temiz *et al.,* 2005).

### 3.4 Surface roughness
### 3.4. Hrapavost površine

The overall surface quality of wood is defined by several properties, with well-recognized importance of surface roughness (Yildiz *et al.,* 2011). However, wood is heterogeneous, anisotropic and fragile material. The roughness of the surface of wood materials depends on many factors such as the anatomical characteristics of wood (vessels, light of cells, width of annual rings, hardness, etc.), conditions of machining process (speed of feed, cutting speed, etc.) and cutting properties (Karagoz *et al.,* 2011). In addition, rough-surfaced wood materials require much more sanding than smooth-surfaced materials resulting in a decrease in the thickness of the material and consequently an increase in sanding losses (Dundar *et al.,* 2008). The evolution of wood surface roughness parameters (*Ra*, *Rz,* and *Rq*) before and after artificial weathering is summarized in Table 4. Before

**Table 3** Color changes of samples before and after artificial weathering
**Tablica 3.** Promjene boje uzorka prije i nakon umjetnog izlaganja vremenskim utjecajima

| **Impregnation and coating types** *Vrste impregnacije i premza* | **Before artificial weathering** *Prije umjetnog izlaganja vremenskim utjecajima* | | | **After 250 h artificial weathering** *Nakon 250 h umjetnog izlaganja vremenskim utjecajima* | | | **After 500 h artificial weathering** *Nakon 500 h umjetnog izlaganja vremenskim utjecajima* | | |
|---|---|---|---|---|---|---|---|---|---|
| | $L_i^*$ | $a_i^*$ | $b_i^*$ | $L_f^*$ | $a_f^*$ | $b_f^*$ | $L_f^*$ | $a_f^*$ | $b_f^*$ |
| Control / *kontrolni uzorci* | 75.69 | 5.87 | 54.58 | 59.02 | 12.29 | 47.37 | 54.58 | 12.12 | 42.52 |
| ATFB + Liquid glass<br>*ATFB + tekuće staklo* | 67.58 | 9.40 | 47.96 | 51.64 | 12.78 | 47.70 | 47.96 | 13.95 | 48.43 |
| APB + Liquid glass<br>*APB + tekuće staklo* | 62.97 | 6.55 | 49.34 | 53.23 | 10.43 | 48.90 | 49.34 | 11.99 | 49.39 |
| BA + Liquid glass<br>*BA + tekuće staklo* | 65.08 | 6.72 | 50.57 | 52.75 | 10.10 | 45.78 | 50.57 | 11.82 | 48.08 |
| Liquid glass<br>*tekuće staklo* | 68.63 | 7.35 | 53.03 | 55.57 | 10.97 | 49.10 | 53.03 | 12.61 | 52.17 |

| | **Total color change after 250 hours** *Ukupna promjena boje nakon 250 h* | | | | **Total color change after 500 hours** *Ukupna promjena boje nakon 500 h* | | | |
|---|---|---|---|---|---|---|---|---|
| | $\Delta L^*$ | $\Delta a^*$ | $\Delta b^*$ | $\Delta E^*$ | $\Delta L^*$ | $\Delta a^*$ | $\Delta b^*$ | $\Delta E^*$ |
| Control / *kontrolni uzorci* | -16.67 | 6.42 | 10.19 | 20.56 | -21.11 | 6.25 | 5.34 | 22.65 |
| ATFB + Liquid glass<br>*ATFB + tekuće staklo* | -15.94 | 3.38 | 8.32 | 18.29 | -19.62 | 4.55 | 9.05 | 22.08 |
| APB + Liquid glass<br>*APB + tekuće staklo* | -9.74 | 3.88 | 9.47 | 14.12 | -13.63 | 5.44 | 9.96 | 17.73 |
| BA + Liquid glass<br>*BA + tekuće staklo* | -12.33 | 3.38 | 9.62 | 15.99 | -14.51 | 5.1 | 11.92 | 19.45 |
| Liquid glass<br>*tekuće staklo* | -13.06 | 3.62 | 12.6 | 18.50 | -15.6 | 5.26 | 15.67 | 22.72 |

**Table 4** Surface roughness of samples before and after artificial weathering
**Tablica 4.** Hrapavost površine uzoraka prije i nakon umjetnog izlaganja klimatskim utjecajima

| **Impregnation and coating types** *Vrste impregnacije i premaza* | **Before artificial weathering** *Prije umjetnog izlaganja vremenskim utjecajima* | | | **After 250 h artificial weathering** *Nakon 250 h umjetnog izlaganja vremenskim utjecajima* | | | **After 500 h artificial weathering** *Nakon 500 h umjetnog izlaganja vremenskim utjecajima* | | |
|---|---|---|---|---|---|---|---|---|---|
| | *Ra* | *Rz* | *Rq* | *Ra* | *Rz* | *Rq* | *Ra* | *Rz* | *Rq* |
| Control / *kontrolni uzorci* | 3.75 | 19.90 | 4.73 | 4.78 | 28.59 | 6.39 | 4.74 | 26.39 | 5.89 |
| ATFB + Liquid glass<br>*ATFB + tekuće staklo* | 0.37 | 2.66 | 0.57 | 0.41 | 2.85 | 0.60 | 0.42 | 2.72 | 0.60 |
| APB + Liquid glass<br>*APB + tekuće staklo* | 0.25 | 1.53 | 0.33 | 0.31 | 1.67 | 0.40 | 0.33 | 1.65 | 0.38 |
| BA + Liquid glass<br>*BA + tekuće staklo* | 0.18 | 1.40 | 0.25 | 0.23 | 1.54 | 0.32 | 0.24 | 1.57 | 0.32 |
| Liquid glass<br>*tekuće staklo* | 0.31 | 2.53 | 0.46 | 0.42 | 2.74 | 0.70 | 0.52 | 2.83 | 0.64 |

| | **Change after 250 hours, %** *Promjena nakon 250 h, %* | | | **Change after 500 hours, %** *Promjena nakon 500 h, %* | | |
|---|---|---|---|---|---|---|
| | *Ra* | *Rz* | *Rq* | *Ra* | *Rz* | *Rq* |
| Control / *kontrolni uzorci* | 27.73 | 43.71 | 35.30 | 26.66 | 32.66 | 24.73 |
| ATFB + Liquid glass<br>*ATFB + tekuće staklo* | 12.53 | 7.18 | 6.06 | 16.00 | 2.61 | 6.06 |
| APB + Liquid glass<br>*APB + tekuće staklo* | 24.77 | 9.24 | 22.77 | 32.57 | 7.86 | 17.21 |
| BA + Liquid glass<br>*BA + tekuće staklo* | 33.33 | 10.32 | 28.00 | 38.88 | 12.56 | 29.86 |
| Liquid glass<br>*tekuće staklo* | 38.70 | 8.69 | 54.34 | 70.96 | 12.25 | 41.30 |

weathering, the untreated (control) specimen had average values of *Ra*, *Rz,* and *Rq* of 3.75, 19.90, and 4.73 respectively. Results showed that the surface roughness values of the untreated (control) group were higher than those of the other treatment groups before and after weathering. Light irradiation degrades mainly the middle lamella, which is located between cell walls to hold the cells together. This degradation increases the roughness of the wood material surface (Tolvaj *et al*., 2014). The increase in wood roughness is also related to sudden changes in moisture (absorption and desorption of water) causing the presence of superficial cracks in addition to the leaching of lignin degraded by natural weathering (Kerber *et al*., 2016). In addition to biotic factors, abiotic factors, such as sunlight, rain, wind, and sandblast, can also affect the surface of wood (Salla *et al*., 2012).

The results obtained in this research are therefore consistent with the literature references, and show that the smoothest surface was obtained for APB-impregnated and LG-coated Scots pine.

## 4 CONCLUSIONS
## 4. ZAKLJUČAK

This research was carried out to improve the weathering performance of Scots pine wood. Different impregnation and finishing combinations resulted in diverse weathering patterns affecting Scots pine wood surface hardness, color changes, and surface roughness. There was a noticeable decrease in surface hardness after the weathering of the non-treated Scots pine samples. However, there was a significant increase in surface hardness after the weathering of impregnated and coated samples. Artificial weathering caused a decrease of Scots pine wood lightness ($\Delta L^*$), with the highest decrease observed in the untreated (control) samples. The best color stabilization after weathering was noticed in APB-impregnated and LG-coated Scots pine. Total color changes were lowest for borates-impregnated and LG-coated Scots pine when compared with only LG-coated wood. All treatment scenarios resulted in positive (less advanced) changes of $\Delta a^*$ and $\Delta b^*$ after weathering. The artificial weathering process caused an increase in surface roughness of control (untreated) and all other treatment groups. Borates-impregnated and LG-coated Scots pine had smoother surfaces than the control wood.

Borates-impregnated and LG-coated Scots pine hardened its surface after weathering. Moreover, such finishing resulted in smoother surfaces than those of the untreated (control) wood. Borates impregnation before LG coating assures better color stability than just coating with liquid glass.

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# An in Vitro Study on Antifungal Properties, Total Polyphenolic Content and Antioxidant Activity of Different Parts of Selected Fruit Trees

## *In vitro* istraživanje protugljivičnih svojstava, ukupnog sadržaja polifenola i antioksidativne aktivnosti različitih dijelova odabranih voćkarica

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**ABSTRACT •** *The purpose of this study was to investigate the antifungal properties, total polyphenolic content and antioxidant activity of sapwood, heartwood and bark parts obtained from three different fruit trees; olive (Olea europaea L.), loquat (Eriobotrya japonica Lindl.) and date plum (Diospyros lotus L.) grown in Turkey. Firstly, the hydrodistillation products (essential oil, hydrosol, hot-water extract) were acquired from these parts of selected trees, and methanol and ethanol extracts of the same samples were also prepared for the analysis. The antifungal activities of essential oil, hydrosol, hot-water extract, ethanol and methanol extracts were determined against wood-rot fungus Coniophora puteana. In addition, polyphenol contents and antioxidant activities of ethanol and methanol extracts were investigated. According to the results, generally, the essential oil of the parts of three fruit trees was found to inhibit the fungal growth. All hot water extracts and hydrosols of these trees showed no antifungal activity against C. puteana. It was found that olive tree had higher antifungal activity than loquat and date plum trees. The methanol extract of loquat bark gave the highest total phenolic content, total flavonoid content and condensed tannin content compared to parts of the other trees. Furthermore, the highest total polyphenol content and antioxidant activity among three fruit trees were determined in ethanol and methanol extracts of the bark and heartwood of loquat tree. Based on these findings, it can be concluded that the parts of studied fruit trees have less or more antifungal and antioxidant activity depending on the experimental parameters, and therefore they can be evaluated as alternative natural antifungal and antioxidant sources.*

***Keywords:*** *antioxidant; antifungal; bark; date plum; loquat; olive; wood*

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**SAŽETAK •** *Cilj ovog rada bio je istražiti protugljivična svojstva, ukupni sadržaj polifenola i antioksidativne aktivnosti bjeljike, srži i dijelova kore drva triju voćkarica koje se uzgajaju u Turskoj: masline (Olea europaea L.), nušpule (Eriobotrya japonica Lindl.) i draguna (Diospyros lotus L.). Najprije su od dijelova odabranih stabala izolirani produkti hidrodestilacije (esencijalna ulja, hidrosol, vodeni ekstrakt), a zatim su za analizu izolirani i ekstrakti u metanolu i etanolu. Protugljivična aktivnost esencijalnog ulja, hidrosola, vodenog ekstrakta te ekstrakata u metanolu i etanolu određena je na primjeru gljive uzročnice smeđe truleži, Coniophora puteana. Osim toga, istraženi su sadržaj polifenola i antioksidativna aktivnost ekstrakta u metanolu i etanolu. Rezultati pokazuju da esencijalna ulja iz dijelova voćkarica usporavaju rast gljiva. Svi vodeni ekstrakti i hidrosol nisu pokazali protugljivično djelovanje na C. puteana. Utvrđeno je da drvo masline ima jaču protugljivičnu aktivnost nego drvo nušpule i drvo draguna. Ekstrakt kore nušpule u metanolu ima najveći ukupni sadržaj fenola, ukupni sadržaj flavonoida i kondenziranog tanina u usporedbi s dijelovima ostalih vrsta drva. Nadalje, za ekstrakt kore i srži drva nušpule u etanolu i metanolu utvrđen je najveći ukupni sadržaj polifenola i antioksidativna aktivnost od svih triju voćkarica. Na temelju tih rezultata može se zaključiti da dijelovi istraživanih voćkarica imaju manje ili više protugljivično i antioksidativno djelovanje, ovisno o parametrima istraživanja te se stoga mogu ocijeniti kao alternativni prirodni izvori protugljivičnih i antioksidativnih sredstava.*

***Ključne riječi:*** *antioksidativno sredstvo; protugljivično sredstvo; kora, dragun; nušpula; maslina; drvo*

## 1 INTRODUCTION
## 1. UVOD

Turkey is characterized by great bioclimatic and geomorphological diversity, and is located at the intersection of three biogeographical regions (Euro-Siberian, Mediterranean and Irano-Turanian) (Noroozi *et al.,* 2019). In Turkey, where forests cover 28.6 % (22.3 million ha) of the land, about 75 fruit species, including 59 temperate zone and 16 subtropical fruit species are commercially grown (FAO, 2018). The main fruits produced in Turkey can be summarized as follows: olive, grape, bananas, kiwi, avocado, figs, pome fruits (apple, pears, medlar, quinces, loquats), citrus fruits (oranges, mandarin, lemons, grapefruits), stone fruits (peaches, plums, apricots, wild apricots, cherries, sour cherries, cornel, oleaster), nuts (almonds, hazelnuts, walnuts, chestnuts, pistachios), and other fruits such as mulberry, pomegranates, strawberries, persimmons, carobs, raspberry, blackberry, blueberry (TÜİK, 2018a).

Olive tree (*Olea europaea* L.) belongs to the *Oleaceae* family, which is a medium-sized family of approximately 25 genera and 688 species, and is a long-lived evergreen tree. It is grown, both in the northern and southern hemispheres, between 30° and 45° latitude, in areas up to 2000 m high in Argentina, in areas below sea level in the Jordan Valley and Israel. Italy, Turkey, Greece, Spain, Cyprus, Portugal, Tunisia, Morocco, Syria, Algeria, France, Libya, former Yugoslavia, Egypt, Jordan and Israel produce about 97 % of the world supply of olives today (CABI, 2019a). Olive tree is famous with its fruit and it has commercially important as the main source for the production of olive oil in the Mediterranean region (Ghanbari *et al.*, 2012).. Turkey has a big potential for olive production and this production shows a tremendous increase every year. According to 2018 data, in Turkey, the total number of olive trees is 177,843.000 and the total production including table olives and oil is 1,500.467 tons (TÜİK, 2018b).

Loquat (*Eriobotrya japonica* (Thunb.) Lindl.) belongs to the *Rosaceae* family, having 92 genera and 2,805 species of herbs, shrubs and trees, and it is a small evergreen tree that occasionally growing up to 10 meters. Loquat is indigenous to south-eastern China and Japan, and generally loquats are found between latitudes of 20° and 35° north and south. In addition, loquats are widely grown across India and South-East Asia, the Mediterranean region (particularly Spain and Turkey), the East Indies, Australia, New Zealand, South Africa and Madagascar (CABI, 2019b). Loquat has a high medicinal value and its various parts have been utilized as traditional medicine. In Chinese folk medicine, the extracts of loquat have been used for the cure of various illnesses such as chronic bronchitis, cough, diabetes, inflammation, and cancer (Liu *et al.*, 2016). According to 2018 data, in Turkey, the total number of loquat trees, including bearing and non-bearing, is 285 thousand (TÜİK, 2018c).

Date plum (*Diospyros lotus* L.) is a deciduous tree that can grow up to 15 meters. It spreads from north Anatolia, Caucasia and Iran to Afghanistan, India, China and Japan (Mamıkoğlu, 2007). It has been proven that its fruit is a natural source of antioxidants (Gao *et al*., 2014) and exhibits hypoglycemic properties (Zhang *et al*., 2018) and anti-tumor-promoting activities (Rauf *et al*., 2016). In addition, all three fruit trees (olive, loquat and date plum) have diffuse-porous wood, paratracheal and apotracheal longitudinal parenchyma cells, and uniseriate and biseriate heterocellular rays (Topaloğlu *et al*., 2019).

As a result of the literature research, it was determined that the antifungal and antioxidant properties of leaves and fruits of the three fruit trees were investigated rather than wood properties of the trees. If these mentioned properties of stem wood and bark of these fruit trees are known, their potential use will increase in different areas. Therefore, the purposes of this study are:

- to determine the antifungal properties of ethanol and methanol extracts and hydrodistillation products such as essential oil, hydrosol and hot-water extract of sapwood, heartwood and bark of fruit trees,
- to determine the total polyphenol contents and antioxidant activity of ethanol and methanol extracts of sapwood, heartwood and bark of these trees.

## 2 MATERIALS AND METHODS
## 2. MATERIJALI I METODE

### 2.1 Sample trees
### 2.1. Uzorci drva

In this study, olive (*Olea europaea* L.), loquat (*Eriobotrya japonica* Lindl.) and date plum (*Diospyros lotus* L.) trees were taken as raw materials. A total of three trees of 30 cm in diameter and 45-50 years of age, with straight bole, no pests and diseases, and showing normal branching, were selected as the experiment materials. Olive and date plum trees were obtained from Artvin city, Yusufeli District (40º53' north latitude - 41º42' east longitude) and loquat tree from Trabzon city, Arsin District (40º55' north latitude - 39º56' east longitude) located in the Black Sea region of Turkey.

### 2.2 Preparation of test samples
### 2.2. Priprema ispitnih uzoraka

After harvesting, the logs were cut from these trees and barks were separated from these logs. These barks and logs were naturally air-dried. Then, each log was sawn into the boards having sapwood and heartwood. These samples were conditioned at 20 °C temperature and 65 % relative humidity until they reached to 12 % moisture content. Air-dried particles of wood were grounded using a laboratory mill (Thomas-Wiley) screened and the samples stayed above 60 mesh were taken and prepared for analysis. All test samples were labeled as bark, sapwood and heartwood. Firstly, the hydrodistillation products (essential oil, hydrosol, hot water extract) of grounded samples were obtained in this study, and also ethanol and methanol extracts of the grounded samples were prepared.

### 2.3 Hydrodistillation products
### 2.3. Produkti hidrodestilacije

Water distillation method which is one of the hydrodistillation methods was used to obtain hydrodistillation products (essential oil, hydrosol, hot water extract) from grounded samples. Nearly 30 g of each grounded sample was taken and separately subjected to hydrodistillation process using a clevenger type apparatus for 3 hours according to the related method (Clevenger, 1928). After this process; essential oil, hydrosol, hot water extract products were obtained and stored at -20 °C in amber vials before analysis. Essential oil, hydrosol, hot water extract of studied materials were used directly. It means the mentioned solutions were not extracted with any solvent.

### 2.4 Ethanol and methanol extracts
### 2.4. Ekstrakti u etanolu i metanolu

For extract preparation, about 5 g of grounded sample was taken and extracted seperately with 40 ml of ethanol and methanol solvents. These were continuously stirred using a shaker (Heidolph, Promax 2020, Schwabach, Germany) for 24 hours. Particles in the mixtures were removed with filter paper (No.4, pore size 20-25 µm) and solutions were filter-sterilized using 0.45 µm hydrophilic polyvinylidene fluoride (PVDF) (Yıldız *et al.,* 2018). A total of 18 ethanol and methanol extracts were prepared from sapwood, heartwood and bark of three fruit trees.

### 2.5 Antifungal properties
### 2.5. Protugljivična svojstva

In this study, antifungal properties of the samples obtained from sapwood, heartwood and bark of fruit trees were tested against brown rot fungus *Coniophora puteana* BAM Ebw. 15, according to the method mentioned in the study by Singh and Tripathi (1999). Firstly, malt extract agar solutions were prepared. Then, 15 ml of malt extract agar and 2 ml of the studied materials (essential oil, hydrosol, hot-water extract, ethanol and methanol extract) were mixed in sterilized petri dishes, separately. After cooling, 5 mm × 5 mm mycelium of *C. puteana* was added to the mixture in every petri dish. Then, all petri dishes were incubated at 22 ±3 °C and 65 ±5 % relative humidity. After that, the entire petri dish diameter (millimeter) and then the fungal mycelium diameter (millimeter) in petri dish were measured at the end of the 3rd, 5th and 7th day using Digimizer Image Analysis Software program version 5.4.1. Fungal growth rate (%) was calculated as a percentage ratio of the mycelium diameter to the entire petri dish diameter. All experiments were replicated three times.

### 2.6 Total polyphenol content and antioxidant activity
### 2.6. Ukupni sadržaj polifenola i antioksidativno djelovanje

In this study, total polyphenol content and antioxidant activity of ethanol and methanol extracts were investigated. Total polyphenol contents were determined by three different assays: total phenolic content (TPC), total flavonoid content (TFC) and condensed tannin content (CTC). Also, Ferric Reducing Antioxidant Power (FRAP) was tested as antioxidant activity.

#### 2.6.1 Determination of total polyphenol content
#### 2.6.1. Utvrđivanje ukupnog sadržaja polifenola

#### 2.6.1.1 Total phenolic content (TPC)
#### 2.6.1.1. Ukupni sadržaj fenola (TPC)

Total phenolic content (TPC) measurements were made according to Folin-Ciocalteu method mentioned by Slinkard and Singleton (1977). This method briefly outlined as follows: 680 µL distilled water, and 400 µl Folin-Ciocalteu reagent (0.5 N), 20 µl various concentrations of gallic acid were prepared and mixed with samples in a tube and vortexed. After 3 min incubation, 400 µl $Na_2CO_3$ 10 % (v/v) solution was added into the tube and vortexed again. Then, the mixture was incubated for 2 hours at 20 °C. After this incubation period, the absorbance of these sample mixtures was determined using an UV-VIS spectrophotometer at 760 nm. The measurement results were expressed as mg gallic acid equivalents (GAE) per g dry weight of the sample.

#### 2.6.1.2 Total flavonoid content (TFC)
#### 2.6.1.2. Ukupni sadržaj flavonoida (TFC)

Total flavonoid content (TFC) measurements of the samples were performed according to the method previously reported by Fukumoto and Mazza (2000). In brief, 0.5 ml sample, 0.1 ml 10 % $Al(NO_3)_3$ and 0.1 ml 1 M $NH_4 \cdot CH_3COO$ were put into test tubes and incubated for 40 min. at room temperature. Then, the absorbances of the samples were measured against a blank at 415 nm. Results of this analysis were expressed as mg quercetin equivalents (QE) per g dry weight of the sample.

#### 2.6.1.3 Condensed tannin content (CTC)
#### 2.6.1.3. Sadržaj kondenziranog tanina (CTC)

Condensed tannin content (CTC) measurements of samples were carried out according to the method reported by Julkunen-Tiitto (1985). Before measurement, 750 µl 4 % vanillin (prepared with MeOH) and various concentrations of 25 µl from extracts of wood samples were mixed. After this step, 375 µl concentrated HCl was added to the mixture. This solution was thoroughly mixed and then incubated for 20 min at room temperature under darkness condition. In this measurement, the absorbance of samples was read at 500 nm against a blank. In order to make the standard curve (0.05–1 mg/ml), (+)-catechin was used as standard. The analysis results were expressed as mg of catechin equivalent (CE)/g of dried sample.

### 2.6.2 Antioxidant activity
### 2.6.2. Antioksidativno djelovanje

#### 2.6.2.1 Ferric reducing antioxidant power (FRAP)
#### 2.6.2.1. Redukcijska antioksidativna snaga željeza (FRAP)

Ferric reducing antioxidant power (FRAP) assay of the samples was carried out as stated by Benzie and Strain (1996) with a slight modification. According to this method, the fresh FRAP reagent was prepared by using the mixture of 300 mM acetate buffer (pH 3.6) with 10 mM, TPTZ solution in 40 mM HCl and 20 mM $FeCl_3$ $6H_2O$ solution. Also, 3ml FRAP reagent was mixed with 100 µl samples and this mixture was incubated at 37 °C for 4 min. The absorbance of the sample was measured at 593 nm against reagent blank containing distilled water. $FeSO_4 \cdot 7H_2O$ was used for measurement as positive control. FRAP values were determined as µmol $FeSO_4 \cdot 7H_2O$ $g^{-1}$ dry weight of the sample.

### 2.7 Statistical analysis
### 2.7. Statistička analiza

Statistical analysis was performed using the SPSS 23.0 version. Analysis of variance (One-Way ANOVA) for all experimental parameters was performed on the basis of the 95 % confidence interval. The mean and standard deviation values of each group were calculated. Duncan's homogeneity test was performed to determine significant differences among parameters.

## 3 RESULTS AND DISCUSSION
## 3. REZULTATI I RASPRAVA

### 3.1 Antifungal activity
### 3.1. Protugljivično djelovanje

Mean values of fungal growth rates (%) of *C. puteana* on solutions of date plum tree parts in petri dishes are presented in Table 1.

Results showed that the essential oil and ethanol extract of both sapwood and heartwood of date plum tree, as well as the essential oil of date plum tree bark completely inhibited fungal growth of *C. puteana* at the end of the seventh day. Hot-water extract and hydrosol of sapwood, heartwood and bark of date plum tree did not show antifungal activity. It was also observed that the methanol extract of all three parts of date plum tree showed no anti-fungal activity after the third day. According to the results of One-Way ANOVA test, there were statistically significant differences between control and test samples ($P \leq 0.05$), control

**Table 1** Fungal growth rates of *C. puteana* on solutions of date plum tree parts in petri dishes (%)
**Tablica 1.** Stupanj rasta gljive *C. puteana* na otopinama dijelova drva draguna u Petrijevim zdjelicama (%)

| **Tree parts**<br>*Dijelovi drva* | **Solutions**<br>*Otopina* | **3rd day**<br>*Treći dan* | **5th day**<br>*Peti dan* | **7th day**<br>*Sedmi dan* |
|---|---|---|---|---|
| Sapwood of date plum tree<br>*bjeljika drva draguna* | Hot-water extract / *vodeni ekstrakt* | 7.52 (1.80)$^{cd}$ | 52.49 (8.13)$^{e}$ | 100.00 (0.00)$^{f}$ |
| | Hydrosol / *hidrosol* | 8.89 (2.62)$^{d}$ | 49.51 (6.21)$^{e}$ | 100.00 (0.00)$^{f}$ |
| | Methanol extract / *ekstrakt u metanolu* | 0.00 (0.00)$^{a}$ | 8.30 (0.42)$^{b}$ | 12.35 (0.49)$^{c}$ |
| | Ethanol extract / *ekstrakt u etanolu* | 0.00 (0.00)$^{a}$ | 0.00 (0.00)$^{a}$ | 0.00 (0.00)$^{a}$ |
| | Essential oil / *esencijalno ulje* | 0.00 (0.00)$^{a}$ | 0.00 (0.00)$^{a}$ | 0.00 (0.00)$^{a}$ |
| Heartwood of date plum tree<br>*srž drva draguna* | Hot-water extract / *vodeni ekstrakt* | 7.91 (1.24)$^{d}$ | 64.58 (9.03)$^{f}$ | 100.00 (0.00)$^{f}$ |
| | Hydrosol / *hidrosol* | 8.26 (0.25)$^{d}$ | 54.21 (5.67)$^{e}$ | 100.00 (0.00)$^{f}$ |
| | Methanol extract / *ekstrakt u metanolu* | 0.00 (0.00)$^{a}$ | 10.91 (1.28)$^{bc}$ | 21.75 (1.76)$^{d}$ |
| | Ethanol extract / *ekstrakt u etanolu* | 0.00 (0.00)$^{a}$ | 0.00 (0.00)$^{a}$ | 0.00 (0.00)$^{a}$ |
| | Essential oil / *esencijalno ulje* | 0.00 (0.00)$^{a}$ | 0.00 (0.00)$^{a}$ | 0.00 (0.00)$^{a}$ |
| Bark of date plum tree<br>*kora drva draguna* | Hot-water extract / *vodeni ekstrakt* | 6.23 (0.03)$^{c}$ | 37.11 (5.25)$^{d}$ | 57.42 (4.97)$^{e}$ |
| | Hydrosol / *hidrosol* | 3.11 (1.41)$^{b}$ | 16.70 (3.42)$^{c}$ | 58.74 (5.07)$^{e}$ |
| | Methanol extract / *ekstrakt u metanolu* | 0.00 (0.00)$^{a}$ | 1.37 (0.28)$^{a}$ | 8.53 (0.37)$^{b}$ |
| | Ethanol extract / *ekstrakt u etanolu* | 0.00 (0.00)$^{a}$ | 1.09 (0.12)$^{a}$ | 3.32 (0.28)$^{a}$ |
| | Essential oil / *esencijalno ulje* | 0.00 (0.00)$^{a}$ | 0.00 (0.00)$^{a}$ | 0.00 (0.00)$^{a}$ |
| Control / *kontrolni uzorci* | | 11.41 (0.58)$^{e}$ | 67.28 (1.81)$^{f}$ | 100.00 (0.00)$^{f}$ |

Values in parenthesis are standard deviations. Different superscript letters in the same column indicate significant difference by Duncan's homogeneity test, $P \leq 0.05$. / *Vrijednosti u zagradama standardne su devijacije. Različita slova u eksponentu u istom stupcu upućuju na značajnu razliku utvrđenu Duncanovim testom, $P \leq 0,05$.*

**Table 2** Fungal growth rates of *C. puteana* on solutions of loquat tree parts in petri dishes (%)
**Tablica 2.** Stupanj rasta gljive *C. puteana* na otopinama dijelova drva nušpule u Petrijevim zdjelicama (%)

| **Tree parts**<br>*Dijelovi drva* | **Solutions**<br>*Otopina* | **3rd day**<br>*Treći dan* | **5th day**<br>*Peti dan* | **7th day**<br>*Sedmi dan* |
|---|---|---|---|---|
| Sapwood of loquat tree<br>*bjeljika drva nušpule* | Hot-water extract / *vodeni ekstrakt* | 8.15 (2.83)$^{e}$ | 57.42 (4.97)$^{e}$ | 100.00 (0.00)$^{g}$ |
| | Hydrosol / *hidrosol* | 7.89 (1.27)$^{de}$ | 59.77 (1.66)$^{e}$ | 100.00 (0.00)$^{g}$ |
| | Methanol extract / *ekstrakt u metanolu* | 0.00 (0.00)$^{a}$ | 0.00 (0.00)$^{a}$ | 0.00 (0.00)$^{a}$ |
| | Ethanol extract / *ekstrakt u etanolu* | 0.00 (0.00)$^{a}$ | 1.98 (0.03)$^{a}$ | 16.88 (2.65)$^{c}$ |
| | Essential oil / *esencijalno ulje* | 0.00 (0.00)$^{a}$ | 45.63 (2.31)$^{d}$ | 54.13 (3.01)$^{d}$ |
| Heartwood of loquat tree<br>*srž drva nušpule* | Hot-water extract / *vodeni ekstrakt* | 9.07 (0.39)$^{e}$ | 74.65 (6.57)$^{g}$ | 100.00 (0.00)$^{g}$ |
| | Hydrosol / *hidrosol* | 4.39 (0.41)$^{b}$ | 34.55 (3.61)$^{bc}$ | 59.77 (1.66)$^{e}$ |
| | Methanol extract / *ekstrakt u metanolu* | 0.00 (0.00)$^{a}$ | 2.05 (0.98)$^{a}$ | 15.03 (1.37)$^{c}$ |
| | Ethanol extract / *ekstrakt u etanolu* | 0.00 (0.00)$^{a}$ | 0.00 (0.00)$^{a}$ | 0.00 (0.00)$^{a}$ |
| | Essential oil / *esencijalno ulje* | 0.00 (0.00)$^{a}$ | 0.00 (0.00)$^{a}$ | 0.00 (0.00)$^{a}$ |
| Bark of loquat tree<br>*kora drva nušpule* | Hot-water extract / *vodeni ekstrakt* | 6.19 (1.68)$^{c}$ | 29.79 (3.12)$^{b}$ | 87.95 (0.08)$^{f}$ |
| | Hydrosol / *hidrosol* | 6.33 (0.99)$^{cd}$ | 38.67 (6.08)$^{c}$ | 55.73 (0.59)$^{d}$ |
| | Methanol extract / *ekstrakt u metanolu* | 0.00 (0.00)$^{a}$ | 1.20 (0.29)$^{a}$ | 10.37 (0.52)$^{b}$ |
| | Ethanol extract / *ekstrakt u etanolu* | 0.00 (0.00)$^{a}$ | 0.00 (0.00)$^{a}$ | 0.00 (0.00)$^{a}$ |
| | Essential oil / *esencijalno ulje* | 0.00 (0.00)$^{a}$ | 0.00 (0.00)$^{a}$ | 0.00 (0.00)$^{a}$ |
| Control / *kontrolni uzorci* | | 11.41 (0.58)$^{f}$ | 67.28 (1.81)$^{f}$ | 100.00 (0.00)$^{g}$ |

Values in parenthesis are standard deviations. Different superscript letters in the same column indicate significant difference by Duncan's homogeneity test, $P \leq 0.05$. / *Vrijednosti u zagradama standardne su devijacije. Različita slova u eksponentu u istom stupcu upućuju na značajnu razliku utvrđenu Duncanovim testom,* $P \leq 0,05$.

**Table 3** Fungal growth rates of *C. puteana* on solutions of olive tree parts in petri dishes (%)
**Tablica 3.** Stupanj rasta gljive *C. puteana* na otopinama dijelova drva masline u Petrijevim zdjelicama (%)

| **Tree parts**<br>*Dijelovi drva* | **Solutions**<br>*Otopina* | **3rd day**<br>*Treći dan* | **5th day**<br>*Peti dan* | **7th day**<br>*Sedmi dan* |
|---|---|---|---|---|
| Sapwood of olive tree<br>*bjeljika drva masline* | Hot-water extract / *vodeni ekstrakt* | 8.14 (2.73)$^{c}$ | 42.01 (1.40)$^{de}$ | 100.00 (0.00)$^{d}$ |
| | Hydrosol / *hidrosol* | 8.65 (3.54)$^{c}$ | 36.62 (4.56)$^{c}$ | 100.00 (0.00)$^{d}$ |
| | Methanol extract / *ekstrakt u metanolu* | 0.00 (0.00)$^{a}$ | 0.00 (0.00)$^{a}$ | 2.78 (1.04)$^{a}$ |
| | Ethanol extract / *ekstrakt u etanolu* | 0.00 (0.00)$^{a}$ | 0.00 (0.00)$^{a}$ | 0.00 (0.00)$^{a}$ |
| | Essential oil / *esencijalno ulje* | 0.00 (0.00)$^{a}$ | 0.00 (0.00)$^{a}$ | 0.00 (0.00)$^{a}$ |
| Heartwood of olive tree<br>*srž drva masline* | Hot-water extract / *vodeni ekstrakt* | 7.68 (1.57)$^{c}$ | 39.26 (8.29)$^{cd}$ | 100.00 (0.00)$^{d}$ |
| | Hydrosol / *hidrosol* | 4.88 (0.72)$^{b}$ | 56.10 (6.84)$^{f}$ | 100.00 (0.00)$^{d}$ |
| | Methanol extract / *ekstrakt u metanolu* | 0.00 (0.00)$^{a}$ | 0.00 (0.00)$^{a}$ | 0.00 (0.00)$^{a}$ |
| | Ethanol extract / *ekstrakt u etanolu* | 0.00 (0.00)$^{a}$ | 0.00 (0.00)$^{a}$ | 0.00 (0.00)$^{a}$ |
| | Essential oil / *esencijalno ulje* | 0.00 (0.00)$^{a}$ | 0.00 (0.00)$^{a}$ | 0.00 (0.00)$^{a}$ |
| Bark of olive tree<br>*kora drva masline* | Hot-water extract / *vodeni ekstrakt* | 1.46 (1.02)$^{a}$ | 26.56 (2.21)$^{b}$ | 38.72 (4.90)$^{b}$ |
| | Hydrosol / *hidrosol* | 5.47 (0.97)$^{b}$ | 46.13 (1.60)$^{e}$ | 66.02 (7.18)$^{c}$ |
| | Methanol extract / *ekstrakt u metanolu* | 0.00 (0.00)$^{a}$ | 0.00 (0.00)$^{a}$ | 0.00 (0.00)$^{a}$ |
| | Ethanol extract / *ekstrakt u etanolu* | 0.00 (0.00)$^{a}$ | 0.00 (0.00)$^{a}$ | 0.00 (0.00)$^{a}$ |
| | Essential oil / *esencijalno ulje* | 0.00 (0.00)$^{a}$ | 0.00 (0.00)$^{a}$ | 0.00 (0.00)$^{a}$ |
| Control / *kontrolni uzorci* | | 11.41 (0.58)$^{d}$ | 67.28 (1.81)$^{g}$ | 100.00 (0.00)$^{d}$ |

Values in parenthesis are standard deviations. Different superscript letters in the same column indicate significant difference by Duncan's homogeneity test, $P \leq 0.05$. / *Vrijednosti u zagradama standardne su devijacije. Različita slova u eksponentu u istom stupcu upućuju na značajnu razliku utvrđenu Duncanovim testom,* $P \leq 0,05$.

samples exhibiting higher fungal growth rates than test samples, especially at the end of the fifth day. According to our results presented in Table 1, the stem wood and bark of date plum have antifungal activity. As a result of the literature research, it was determined that the bioactive properties of fruits, leaves and roots of date plum tree were investigated, while those of stem wood were not. Uddin *et al*. (2013) investigated antimicrobial activities of extracted oil of *D. lotus* roots and Uddin *et al.* (2014) examined anti-nociceptive, anti-inflammatory and sedative activities of the extracts of *D. lotus* roots.

Mean values of fungal growth rates (%) of *C. puteana* on solutions of loquat tree parts in petri dishes are presented in Table 2.

The ethanol extract and essential oil of both heartwood and bark of loquat tree, as well as methanol extract of sapwood of loquat tree completely inhibited fungal growth of *C. puteana* at the end of the seventh day. The hydrosol and hot-water extract of sapwood, heartwood and bark of loquat tree did not show antifungal activity. Methanol extract of heartwood and bark of loquat tree showed no antifungal activity after the third day. According to the results of One-Way ANOVA test, there were statistically significant differences between control and test samples ($P \leq 0.05$). The hot-water extract of heartwood of loquat tree, which had the highest growth rate, was the most ineffective against *C. puteana*, especially compared to the control sample on the fifth day.

Mean values of fungal growth rates (%) of *C. puteana* on solutions of olive tree parts in petri dishes are given in Table 3.

Results showed that the ethanol and methanol extracts and the essential oil of heartwood and bark of olive tree, as well as the essential oil and ethanol extract of sapwood of olive tree had the best antifungal activity by completely inhibiting fungal growth of *C. puteana*. Methanol extract of sapwood of olive tree extract did not entirely prevent the fungal growth. Hot-water and hydrosol of all three parts of olive tree showed no antifungal activity against *C. puteana* at the end of the seventh day. After evaluating the results of antifungal activities obtained in this study for all three fruit trees species, it can be concluded that, generally, all three parts of olive tree showed better antifungal activity compared to loquat and date plum wood species.

There are many studies in the literature related to the antifungal activity of different parts of various wood species against wood-rot fungi. Fidah *et al*. (2016) studied the antifungal activity of *Cedrus atlantica* essential oil against four wood-decaying fungi (*Gloeophyllum trabeum, Oligoporus placenta, Coniophora puteana* and *Trametes versicolor*). They found that the essential oil of *Cedrus atlantica* sawdust had important antifungal activity against the four wood-destroying fungi. Mihara *et al*. (2005) found that the methanol extracts of *Acacia auriculiformis* showed strong antifungal effects on the growth of wood rotting fungi (*Phellinus noxius* and *Phellinus badius*), while the extracts of *Acacia mangium* had no effect on the growth of *Phellinus noxius* and little inhibition of *Phellinus badius.* Ateş *et al.* (2015) examined the antifungal activities of different parts of juniper (*Juniperus foetidissima*) and olive (*Olea europaea* var. *sylvestris)* wood against *Pleurotus ostreatus* mycelium. They found that the extractives (concentration of 500 ppm) of heartwood and sapwood of juniper and olive inhibited (80 %) the growth of *Pleurotus ostreatus* mycelium. Tümen *et al*. (2013), determined the antifungal activity of hexane and ethanol extracts of heartwood samples of three *Juniperus* species (*Juniperus virginiana*, *Juniperus occidentalis,* and *Juniperus ashei*) against four species of wood-rot fungi (*Gloeophyllum trabeum, Postia placenta, Trametes versicolor, Irpex lacteus*). It was found that the hexane extracts had lower antifungal activity than the ethanol extracts, and the extracts of juniper were more effective especially against white-rot fungi compared to brown-rot fungi. Özgenç *et al.* (2017) reported that extracts of some wood barks including *Pinus pinaster* L., *Casuarina equisetifolia* L., *Acacia mollissima* L., *Pinus brutia* Ten., *Abies nordmanniana* showed strong antifungal activity against brown-rot fungus; *Coniophora puteana*, and white-rot fungus; *Trametes versicolor*. They also indicated that antifungal activity rate showed an increase with increasing the concentration of bark extract solutions. Hosseinihashemi *et al.* (2015) investigated the antioxidant and antifungal properties of heartwood, bark and leaf of different fractions of *Robinia pseudoacacia,* and they reported that ethyl acetate fraction of bark extract showed the highest antifungal activity against wood-rot fungi *Trametes versicolor*.

### 3.2 Total polyphenol content and antioxidant activity

### 3.2. Ukupni sadržaj polifenola i antioksidativno djelovanje

Results of total polyphenol content (TPC, TFC, CTC) and antioxidant activity (FRAP) of ethanol and methanol extracts of the parts of date plum, loquat and olive trees are given in Table 4.

As can be seen in Table 4, the highest TPC value was determined for methanol extract of loquat bark with 14.998 mg GAE/g and the lowest for ethanol extract of olive tree sapwood with 0.543 mg GAE/g. Among all methanol and ethanol extracts obtained from different parts of three fruit trees, it was found that the ethanol extracts of barks of date plum trees with 8.866 mg GAE/g and ethanol extracts of loquat bark with 8.494 mg GAE/g had high TPC values. Yıldız *et al.* (2018) reported that the bark of *Acacia dealbata* had the highest total phenolic content. Ateş *et al.* (2015) determined TPC values as the antioxidant activities of heartwood and sapwood of juniper (*Juniperus foetidissima*) and olive (*Olea europaea* var. *sylvestris*). They reported that the highest TPC value was obtained from methanol extract of *Juniperus foetidissima* heartwood. In another study, Hamad *et al.* (2019) reported that the total phenolic content of bark extractives of oak and beech was 48 mg/g and 42.04 mg/g, respectively. Gao *et al.* (2007) investigated the antioxidant activity of methanol extract of sapwood, heartwood, and inner and outer bark of Port Orford cedar. The highest TPC value was detected in the inner bark and the lowest TPC value was observed in the heartwood. In addition, TPC of extracts prepared with different solvents (water, methanol, ethanol, 50 % ethanol, 50 % methanol, 70 % acetone) of *Bridelia retusa* Spreng. stem bark ranged between 1.6-7.6 mg GAE (Tatiya *et al.*, 2011).

As seen in Table 4, the total flavonoid content values were not found in sapwood and heartwood of fruit trees, while TFC values were only found in barks of all studied trees except for ethanol extract of date plum tree and methanol extract of olive tree. The highest TFC was calculated in methanol extract of loquat bark as 0.173 mg QE/g. However, in this study, TFC values were found lower than the results reported in the literature due to the low concentration of the prepared solvent. Ateş *et al.* (2015) investigated TFC values for both heartwood and sapwood of juniper and olive. According to their results, the lowest TFC was found for

**Table 4** Total polyphenol content and antioxidant activity of ethanol and methanol extracts of parts of date plum, loquat and olive trees
**Tablica 4.** Ukupni sadržaj polifenola i antioksidativno djelovanje ekstrakta u etanolu i metanolu iz dijelova drva draguna, nušpule i masline

| **Sample** *Uzorak* | **Part of tree** *Dio drva* | **Solvent** *Otapalo* | **TPC** mg GAE/g | **TFC** mg QE/g | **CTC** mg CE/g | **FRAP** $\mu$molFeSO$_{4.}$7H$_2$O/g |
|---|---|---|---|---|---|---|
| Date plum *dragun* | Sapwood *bjeljika* | E | 1.282 (0.091)$^{ab}$ | - | - | 60.134 (1.344)$^{c}$ |
| | | M | 3.789 (0.005)$^{d}$ | - | - | 67.475 (0.122)$^{e}$ |
| | Heartwood *srž* | E | 1.139 (0.001)$^{ab}$ | - | 0.048 (0.001)$^{a}$ | 58.673 (0.296)$^{c}$ |
| | | M | 3.413 (0.017)$^{cd}$ | - | 0.275 (0.001)$^{b}$ | 88.233 (1.222)$^{f}$ |
| | Bark *kora* | E | 8.866 (0.406)$^{f}$ | - | - | 230.376 (6.166)$^{k}$ |
| | | M | 3.518 (0.003)$^{cd}$ | 0.0002 (0.001)$^{a}$ | 0.131 (0.001)$^{a}$ | 158.890 (0.655)$^{i}$ |
| | | | | | | |
| Loquat *nušpula* | Sapwood *bjeljika* | E | 1.630 (0.045)$^{ab}$ | - | - | 62.186 (1.279)$^{cd}$ |
| | | M | 2.448 (0.015)$^{bc}$ | - | - | 58.430 (2.392)$^{c}$ |
| | Heartwood *srž* | E | 5.264 (0.056)$^{e}$ | - | 2.946 (0.031)$^{d}$ | 97.994 (3.925)$^{g}$ |
| | | M | 6.366 (2.981)$^{e}$ | - | 4.401 (0.018)$^{e}$ | 122.141 (0.698)$^{h}$ |
| | Bark *kora* | E | 8.494 (0.478)$^{f}$ | 0.0003 (0.000)$^{a}$ | 6.815 (0.060)$^{f}$ | 307.791 (0.531)$^{m}$ |
| | | M | 14.998 (0.061)$^{g}$ | 0.173 (0.001)$^{c}$ | 7.703 (0.292)$^{g}$ | 299.251 (5.987)$^{l}$ |
| | | | | | | |
| Olive *maslina* | Sapwood *bjeljika* | E | 0.543 (0.008)$^{a}$ | - | - | 43.522 (0.009)$^{b}$ |
| | | M | 1.647 (0.001)$^{ab}$ | - | 0.269 (0.005)$^{b}$ | 66.032 (0.887)$^{de}$ |
| | Heartwood *srž* | E | 0.774 (0.002)$^{a}$ | - | - | 45.276 (0.089)$^{b}$ |
| | | M | 1.352 (0.185)$^{ab}$ | - | - | 90.075 (0.311)$^{f}$ |
| | Bark *kora* | E | 1.202 (0.081)$^{ab}$ | 0.018 (0.000)$^{b}$ | 0.301 (0.027)$^{b}$ | 31.310 (0.789)$^{a}$ |
| | | M | 0.616 (0.004)$^{a}$ | - | 1.132 (0.001)$^{c}$ | 59.283 (0.512)$^{c}$ |

TPC – total phenolic content, TFC – total flavonoid content, CTC – condensed tannin content, FRAP – ferric reducing antioxidant power, E – ethanol, M – methanol. Values in parenthesis are standard deviations. Different superscript letters in the same column indicate significant difference by Duncan's homogeneity test, $P \leq 0.05$. / *TPC – ukupni sadržaj fenola, TFC – ukupni sadržaj flavonoida, CTC – sadržaj kondenziranog tanina, FRAP – redukcijska antioksidativna snaga željeza, E – etanol, M – metanol. Vrijednosti u zagradama standardne su devijacije. Različita slova u eksponentu u istom stupcu upućuju na značajnu razliku utvrđenu Duncanovim testom, P≤0,05.*

methanol extract of olive heartwood, while the highest TFC was found for ethanol extract of olive sapwood.

Results of samples of the condensed tannin content (CTC) are presented in Table 4. According to these results, it can be concluded that the methanol extracts of loquat bark had the highest CTC value (7.703 mg CE/g), and the second highest CTC value (6.815 mg CE/g) was determined in ethanol extract of loquat bark. In this study, CTC values of methanol extracts of studied tree samples were found higher than those of ethanol extracts. In a study performed by Yıldız *et. al* (2018), the condensed tannin content of methanol extracts of sapwood, heartwood and bark of *Acacia dealbata* was determined as 0.620, 4.473, 4.789 mg CE/g, respectively.

FRAP, used in this study, is one of the most widely used antioxidant methods. FRAP activities of the studied methanol and ethanol extracts ranged between 31.310 and 307.791 µmol FeSO$_{4.}$7H$_2$O/g. While the ethanol extract of loquat bark had the highest antioxidant activity, the ethanol extract of olive bark had the lowest antioxidant activity. Comparing FRAP values for different parts of three fruit trees, it was seen that especially heartwood and bark of loquat tree showed higher values than those of date plum and olive tree. According to the results of One-Way ANOVA test, there were statistically significant differences ($P \leq 0.05$) among experimental groups for TPC, TFC, CTC, FRAP. Furthermore, taking into consideration all the results (TPC, TFC, CTC, FRAP), it can be concluded that, generally, ethanol and methanol extracts of bark and heartwood of loquat had higher values. Some different trends between our results and other results reported in the literature can likely be attributed to various parameters such as different concentration and extraction methods, environmental factors, etc. Based on these results, it was found that all the studied trees have more or less potential to be evaluated as antioxidant and antifungal activity source. Therefore, it is recommended that their potentials can be increased by trying high concentrations and different methods.

## 4 CONCLUSIONS
## 4. ZAKLJUČAK

Important findings of this comparative study carried out for three fruit trees can be summarized as follows:

Generally, essential oil of all fruit trees inhibited the fungal growth rate of *C. puteana*.

All studied hot water extracts and hydrosols showed no antifungal activity against *C. puteana.*

Olive tree showed higher antifungal activity compared to loquat and date plum trees.

The highest total phenolic content (TPC), total flavonoid content (TFC), condensed tannin content (CTC) and antioxidant activity (FRAP) were found in ethanol and methanol extracts of bark and heartwood of loquat. The highest TPC, TFC and CTC were observed in methanol extract of loquat bark.

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# Lin Laboratorij za ispitivanje namještaja i dijelova za namještaj

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# Evaluation of *Rhododendron Luteum* and *Rhododendron Ponticum* in Pulp and Paper Production

## Procjena mogućnosti uporabe biljaka *Rhododendron luteum i Rhododendron ponticum* kao sirovine za proizvodnju celuloze i papira

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**ABSTRACT •** *In this study, Rhododendron luteum and Rhododendron ponticum were evaluated as raw material for pulp and paper production. 12 different sodium borohydride ($NaBH_4$) added cooking trials were performed for each sample and kraft method was used for pulp production. Pulp properties, such as yield, kappa number and viscosity, and physical properties, such as breaking length and burst index, were determined for each trial. Besides, the effects of active alkali and $NaBH_4$ on the pulp and paper properties were also examined. Optimum cooking conditions were obtained by using 18 % active alkali for $NaBH_4$-free cooking experiments and 0.5 % NaBH4 and 18 % active alkali for NaBH4-added cooking experiments. In $NaBH_4$-added pulping condition, the screened yield, kappa number and viscosity of R. luteum were found to be 43.4 %, 40.1 and 949 $cm^3/g^1$, respectively. The respective values for R. ponticum were 41.9 %, 44.5 and 885 $cm^3/g^1$. The screened yields of R. luteum and R. ponticum increased by about 2.8 % and 5.3 %, respectively, with 5 % addition of $NaBH_4$ compared to $NaBH_4$-free cooking experiments. Furthermore, with the addition of $NaBH_4$, the kappa numbers decreased while the viscosity increased. The physical properties of the produced papers were also improved by using $NaBH_4$ in cooking liquor. According to the obtained results, it was found that R. luteum and R. ponticum species can be evaluated for pulp and paper production.*

***Keywords:*** *R. luteum; R. ponticum; pulp; paper; $NaBH_4$*

**SAŽETAK •** *U ovom je radu istražena mogućnost uporabe biljaka Rhododendron luteum i Rhododendron ponticum kao sirovine za proizvodnju celuloze i papira. Za svaki uzorak provedeno je 12 različitih ispitivanja kuhanja s natrijevim borhidridom ($NaBH_4$), a celuloza je proizvedena kraft postupkom. Za svako ispitivanje određena su svojstva celuloze poput prinosa, kappa broja i viskoznosti, te fizička svojstva kao što su duljina lomljenja i indeks pucanja papira. Osim toga, ispitani su učinci aktivne lužine i $NaBH_4$ na svojstva celuloze i papira. Optimalni uvjeti kuhanja postignuti su upotrebom 18 % aktivne lužine za eksperimentalno kuhanje bez $NaBH_4$ i upotrebom 0,5 % $NaBH_4$ i 18 % aktivne lužine za eksperimentalno kuhanje s dodatkom $NaBH_4$. U proizvodnji celuloze iz biljke R. luteum s dodatkom $NaBH_4$ utvrđeno je da prinos prosijavanja iznosi 43,4 %, da je kappa broj 40,1, a viskoznost*

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*949 $cm^3/g$, dok su vrijednosti za celulozu dobivenu iz biljke R. ponticum redom 41,9 %, 44,5 i 885 $cm^3/g$. Prinos prosijavanja biljaka R. luteum i R. ponticum uz dodatak 5 % $NaBH_4$ povećao se oko 2,8 % i 5,3 % u usporedbi s eksperimentalnim kuhanjem bez dodatka $NaBH_4$. Nadalje, uz dodatak $NaBH_4$ smanjuju se kappa brojevi, a viskoznosti se povećavaju. Fizička svojstva proizvedenih papira također se poboljšavaju dodavanjem $NaBH_4$ tekućini za kuhanje. Iz dobivenih je rezultata utvrđeno da se biljke R. luteum i R. ponticum mogu upotrebljavati u proizvodnji celuloze i papira.*

***Ključne riječi:*** *R. luteum; R. ponticum; celuloza; papir; $NaBH_4$*

## 1 INTRODUCTION
## 1. UVOD

In the second half of the twentieth century, especially with the increase of world population and the increase in demand for forest products due to rapid developments in technology, the decrease in forest areas led the forest products industry to search for raw materials as an alternative to wood (Oner and Aslan, 2002; Tutus *et al.*, 2011). Non-wood and new wood products are one of the best alternative raw materials (Kaldor, 1992; Comlekcioglu *et al.*, 2016). Due to the rapid increase in paper consumption in recent years, alternative fibrous materials have gained importance in the pulp and paper industry (Daud and Law, 2011). The search for new wood raw materials still continues in the world.

Rhododendron species, a valuable ornamental shrub, have been cultivated since the 18th century. These species belong to the heath or Ericaceae family. Rhododendron, with about 1000 different species, extends from Southwest Asia to New Guinea. Some Rhododendron species are large species with a tree-type growth habit. It is evergreen or rarely deciduous. Turkish rhododendrons grow naturally up to 3000 m above the sea level. One of the most remarkable Rhododendron species in moist forest formations covering the northern coast of Turkey is *R. luteum* and the most common Rhododendron is *R. ponticum*. Fiber length, fiber width, lumen diameter and cell wall thicknesses of *R luteum and R. ponticum* are approximately 0.9-1.0 mm, 17-19 µm, 7-9 µm and 3-4 µm, respectively. Besides, these species have high holocellulose (77-80 %) and alpha cellulose (47-48 %) contents, which are important carbohydrates for pulp and paper industry (Birinci, 2008; Camlibel, 2008).

Although they appear on the northern slopes of the mountains on the Black Sea coast, there are more species in the Eastern Black Sea Region (Avci, 2004). In many studies, Rhododendron species have generally been evaluated for the production of medium density fiberboard (MDF) (Akgul *et al.*, 2012; Akgul and Camlibel, 2008; Ayrilmis *et al.*, 2014). Besides, some studies have determined the chemical composition of some Rhododendron species (Birinci, 2008; Shrestha and Budhathoki, 2012).

The kraft method is the most commonly used method for obtaining pulp suitable for papermaking. Increasing the pulp yield in pulp production is very important for the pulp and paper industry in terms of cost and economy. Modifying the kraft cooking process, e.g. by adding $NaBH_4$, is a way of increasing the pulp yield (Courchene, 1998; Tutus and Eroglu, 2003; Tutus and Eroglu, 2004; Tutus and Usta 2004; Hafizoglu and Deniz, 2007; Istek and Gonteki, 2009, Akgul *et al.*, 2018). The end groups of carbohydrates are protected from peeling reactions by using $NaBH_4$ as a catalyst in cooking processes (Istek and Ozkan, 2008; Tutus and Cicekler, 2016).

There is little information about the use of Rhododendron species in wood-based industries and there is no study on their use in pulp and paper production. The aim of this study was to evaluate *Rhododendron luteum* and *Rhododendron ponticum* stalks for pulp and paper production, using Kraft-$NaBH_4$ cooking methods.

## 2 MATERIALS AND METHODS
## 2. MATERIJALI I METODE

*R. luteum* and *R. ponticum* species were obtained from Akdamar Village of Akçaabat district of Trabzon province in Turkey. The chemical compositions of *R. luteum* and *R. ponticum* were determined in a previous study (Birinci, 2008) and are presented in Table 1 with some wood and non-wood species.

According to Table 1, the chemical composition of Rhododendron species is similar to that of hardwoods species. Rhododendron species show similarities among themselves. However, the lignin content of *R. ponticum* was higher than that of *R. luteum*. Due to their high holocellulose contents, it was concluded that Rhododendron species were considered suitable for pulp and paper production.

The samples were chipped and air dried. 12 cooking trials were applied on each species, using Kraft-$NaBH_4$ method given in Table 2 in order to determine optimum pulping conditions.

Cooking trials of the species were applied in a rotary digester with a 15-liter capacity and high pressure resistant. After cooking processes, the pulps were washed with tap water until the black liquor was removed, and the washed pulps were transferred to 0.15 mm slotted screen. Screened pulp and screen reject yields were calculated after the screening process. The kappa numbers and viscosity of the screened pulps were determined according to TAPPI T 236 om-13 and TAPPI T 230 om-08, respectively (Anonymous, 1998).

The pulps were beaten to 25 ±3 °SR (Schopper Riegler) freeness level in the hollander beater according to TAPPI T 200 sp-96. Ten test papers were produced from the pulps obtained from each cooking trial. The breaking lengths and burst indices of the papers

**Table 1** Chemical composition of *R. luteum*, *R. ponticum* and some wood and non-wood species
**Tablica 1.** Kemijski sastav biljaka *R. luteum*, *R. ponticum* i nekih drvnih i nedrvnih vrsta

| **Chemical components**<br>*Kemijske sastavnice* | ***R. luteum* (Birinci, 2008)** | ***R. ponticum* (Birinci, 2008)** | ***A. membranaceus* (Tutus *et al.*, 2014)** | ***Fagus orientalis* (Tank, 1978)** | ***R. pseudoacacia* (Kirci, 1987)** | ***P. brutia* (Tutus *et al.*, 2012)** | **Wheat straw (Tutus and Cicekler, 2016)** |
|---|---|---|---|---|---|---|---|
| Holocellulose, %<br>*holoceluloza,* % | 78 | 77 | 77 | 79 | 82 | 79 | 73 |
| Alpha cellulose, %<br>*alfa-celuloza,* % | 48 | 47 | 50 | 42 | 52 | 49 | 39 |
| Lignin, % / *lignin,* % | 31 | 34 | 24 | 23 | 21 | 28 | 18 |
| Ash, % / *pepeo,* % | 0.41 | 0.42 | 5.50 | 0.61 | 0.55 | 0.48 | 7.8 |
| Extractives, %<br>*ekstraktivi,* % | 2.02t | 2.12t | 5.60t | 1.50a | 6.23a | 7.65t | 5.1t |
| %1 NaOH, % | 22.2 | 26.1 | 29.4 | 15.6 | 22.10 | 14.49 | 43.7 |
| Hot water, %<br>*vruća voda,* % | 7.15 | 8.07 | 8.40 | - | - | 2.19 | 14.6 |
| Cold water, %<br>*hladna voda,* % | 1.14 | 0.88 | 7.20 | 1.92 | 8.06 | 1.14 | 11.5 |

*t – toluene-acetone-ethanol extraction, a – alcohol-benzene extraction / **t – ekstrakcija u toluen-acetonu i etanolu, a – ekstrakcija u alkohol-benzenu*

were determined according to TAPPI T 494 om-01 and TAPPI T 403 om-91, respectively (Anonymous, 1998). In order to determine the effects of active alkali and $NaBH_4$, the optimum cooking parameters were used.

**Table 2** Pulping conditions of *R. luteum* and *R. ponticum*
**Tablica 2.** Uvjeti dobivanja celuloze od biljaka *R. luteum* i *R. ponticum*

| **Pulping condition**<br>*Uvjeti proizvodnje celuloze* | **Unit**<br>*Jedinica* | **Value**<br>*Vrijednost* |
|---|---|---|
| Active alkali / *aktivna lužina* | % | 18, 20, 22 |
| Sulfidity / *sulfidnost* | % | 24 |
| $NaBH_4$ charge / *udio* $NaBH_4$ | % | 0, 0.3, 0.5, 0.7 |
| Cooking temperature<br>*temperatura kuhanja* | °C | 160 |
| Time to maximum temperature<br>*vrijeme postizanja najveće temperature* | min | 40 |
| Time at maximum temperature<br>*vrijeme na najvećoj temperaturi* | min | 90 |
| Liquor to raw material ratio<br>*omjer otapala i sirovine* | l/kg | 4/1 |

## 3 RESULTS AND DISCUSSION
## 3. REZULTATI I RASPRAVA

The yield and chemical properties of *R. luteum* and *R. ponticum* pulps used in this study are presented in Table 3.

The screened yield and viscosity of *R. ponticum* was found lower than that of *R. luteum* by about 5.6 % and 15.5 %, respectively. The kappa numbers of *R. ponticum* and *R. luteum* were determined as 49.24 and 45.74. According to these results, *R. luteum* is more suitable for pulping production than *R. ponticum*. The effects of $NaBH_4$ on the properties of Rhododendron pulp are shown in Figure 1.

With the addition of $NaBH_4$ to the cooking liquor, the yield and viscosity of the pulps increased and kappa numbers decreased. $NaBH_4$ was effective on the screened yields as it had the ability to stop peeling reactions occurring in cellulose chains (Istek and Ozkan, 2008; Tutus and Cicekler, 2016). $NaBH_4$ removes the lignin from the pulp i.e. selective lignin delignification occurs, while increasing the viscosity by preserving

**Table 3** Screened yields, kappa numbers and viscosities of *R. luteum* and *R. ponticum* pulps
**Tablica 3.** Prinos prosijavanja, *kappa* brojevi i viskoznosti celuloze dobivene od biljaka *R. luteum* i *R. ponticum*

| **Cooking No**<br>*Broj kuhanja* | **Active alkali, %**<br>*Aktivna lužina, %* | **$NaBH_4$, %** | **Screened yield, %**<br>*Prinos prosijavanja, %* | | **Kappa No**<br>*Kappa broj* | | **Viscosity, $cm^3/g$**<br>*Viskoznost,* $cm^3/g$ | |
|---|---|---|---|---|---|---|---|---|
| | | | *R. luteum* | *R. ponticum* | *R. luteum* | *R. ponticum* | *R. luteum* | *R. ponticum* |
| 1 | 18 | 0 | 42.15 | 39.78 | 45.74 | 49.24 | 895 | 832 |
| 2 | 20 | 0 | 41.78 | 38.50 | 42.16 | 46.78 | 860 | 800 |
| 3 | 22 | 0 | 41.05 | 37.95 | 41.84 | 43.18 | 838 | 786 |
| 4 | 18 | 0.3 | 43.03 | 41.50 | 42.06 | 46.56 | 928 | 870 |
| 5 | 20 | 0.3 | 42.55 | 41.00 | 39.60 | 44.22 | 912 | 854 |
| 6 | 22 | 0.3 | 41.83 | 40.03 | 37.78 | 42.76 | 886 | 808 |
| 7 | 18 | 0.5 | 43.35 | 41.87 | 40.10 | 44.48 | 949 | 885 |
| 8 | 20 | 0.5 | 42.91 | 41.21 | 38.88 | 41.98 | 924 | 868 |
| 9 | 22 | 0.5 | 42.00 | 40.77 | 36.56 | 40.06 | 902 | 829 |
| 10 | 18 | 0.7 | 43.78 | 42.18 | 38.98 | 42.02 | 988 | 902 |
| 11 | 20 | 0.7 | 43.25 | 41.65 | 37.10 | 39.48 | 939 | 880 |
| 12 | 22 | 0.7 | 42.60 | 41.23 | 34.24 | 37.98 | 918 | 840 |

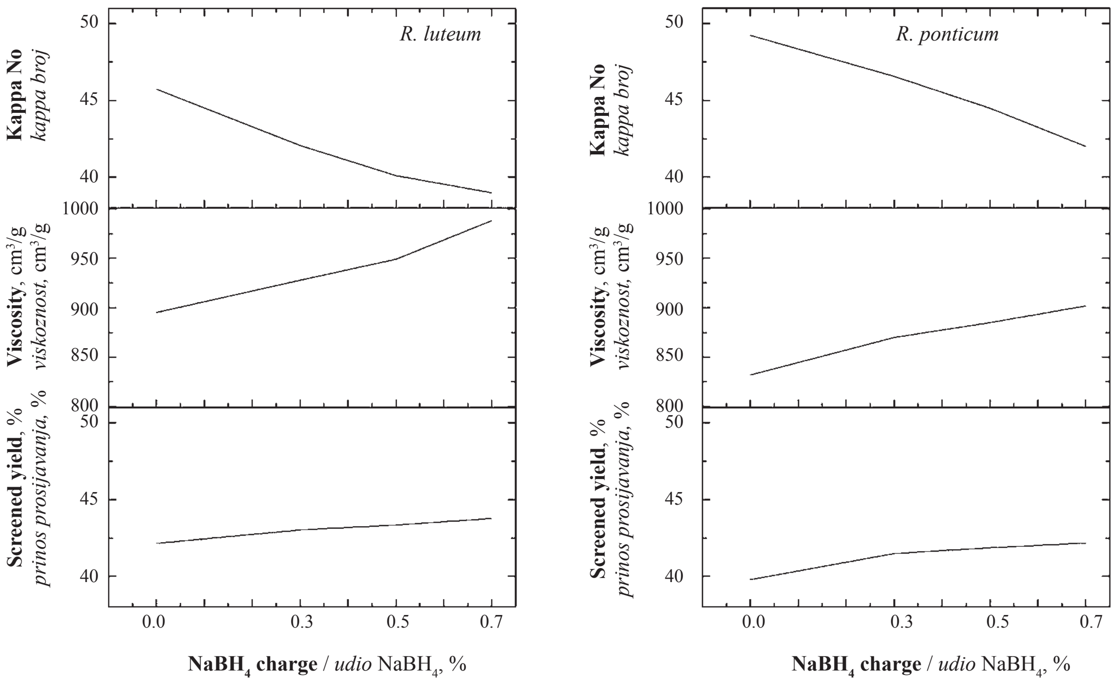

**Figure 1** Effects of $NaBH_4$ charge on yield, kappa number and viscosity of Rhododendron pulps
**Slika 1.** Utjecaj udjela $NaBH_4$ na prinos, *kappa* broj i viskoznost celuloze od rododendrona

carbohydrates (Tutus, 2008). It also prevents shortening of cellulose and hemicellulose chains as it prevents damage to carbohydrates. In this way, the viscosity of the pulps is increased and strength pulps can be obtained. Recent studies have shown a decrease in the kappa numbers of the pulps obtained with the addition of $NaBH_4$ to the cooking experiments (Copur and Tozluoglu, 2008; Gulsoy and Eroglu, 2011; Istek and Gonteki, 2009). With the addition of 0.5 % $NaBH_4$, the screened pulp yields of *R. luteum* and *R. ponticum* pulps increased by approximately 2.8 % (from 42.15 % to 43.35 %) and 5.3 % (from 39.78 % to 41.87 %), while kappa numbers decreased by 12.3 % and 9.8 %, respectively. By keeping the active alkali high, the rate of depolymerization of carbohydrates during cooking increases. Therefore, both yield and viscosity decrease. The screened yields, kappa numbers and viscosity of *R. luteum* and *R. ponticum* pulps decreased by increasing the active alkali from 18 % to 22 %. Many studies have reported that the increase of active alkali decreases the yield, kappa number and viscosity (Lopez et al., 2000; Yue et al., 2016; Zhai and Zhou, 2014).

The breaking lengths and burst indices of *R. luteum* and *R. ponticum* papers used in this study are given in Table 4.

According to Table 4, the breaking lengths and burst indices of the *R. ponticum* papers were found to be higher than those of *R. luteum*. It is clearly seen that the physical properties of the papers improve with the increase of active alkali and $NaBH_4$ ratios (Figure 2).

Many studies have reported that boron compounds prevent peeling reaction during cooking, resulting in less damage to carbohydrates and therefore improved physical and optical properties of the papers

**Table 4** Breaking lengths and burst indices of *R. luteum* and *R. ponticum* papers
**Tablica 4.** Duljina lomljenja i indeks pucanja papira proizvedenog od biljaka *R. luteum* i *R. ponticum*

| **Cooking No** *Broj kuhanja* | **Active alkali, %** *Aktivna lužina, %* | **$NaBH_4$, %** | **Breaking length, km** *Duljina lomljenja,* km | | **Burst index, kPa·m²·g⁻¹** *Indeks pucanja,* kPa·m²·g⁻¹ | |
|---|---|---|---|---|---|---|
| | | | *R. luteum* | *R. ponticum* | *R. luteum* | *R. ponticum* |
| 1 | 18 | 0 | 2.21 | 2.88 | 1.90 | 2.10 |
| 2 | 20 | 0 | 2.29 | 2.91 | 1.98 | 2.25 |
| 3 | 22 | 0 | 2.32 | 3.02 | 2.06 | 2.31 |
| 4 | 18 | 0.3 | 2.35 | 2.98 | 2.00 | 2.28 |
| 5 | 20 | 0.3 | 2.40 | 3.05 | 2.15 | 2.45 |
| 6 | 22 | 0.3 | 2.52 | 3.09 | 2.30 | 2.54 |
| 7 | 18 | 0.5 | 2.57 | 3.07 | 2.17 | 2.43 |
| 8 | 20 | 0.5 | 2.65 | 3.10 | 2.30 | 2.53 |
| 9 | 22 | 0.5 | 2.79 | 3.19 | 2.45 | 2.62 |
| 10 | 18 | 0.7 | 2.87 | 3.15 | 2.32 | 2.51 |
| 11 | 20 | 0.7 | 3.01 | 3.32 | 2.54 | 2.68 |
| 12 | 22 | 0.7 | 3.13 | 3.51 | 2.66 | 2.87 |

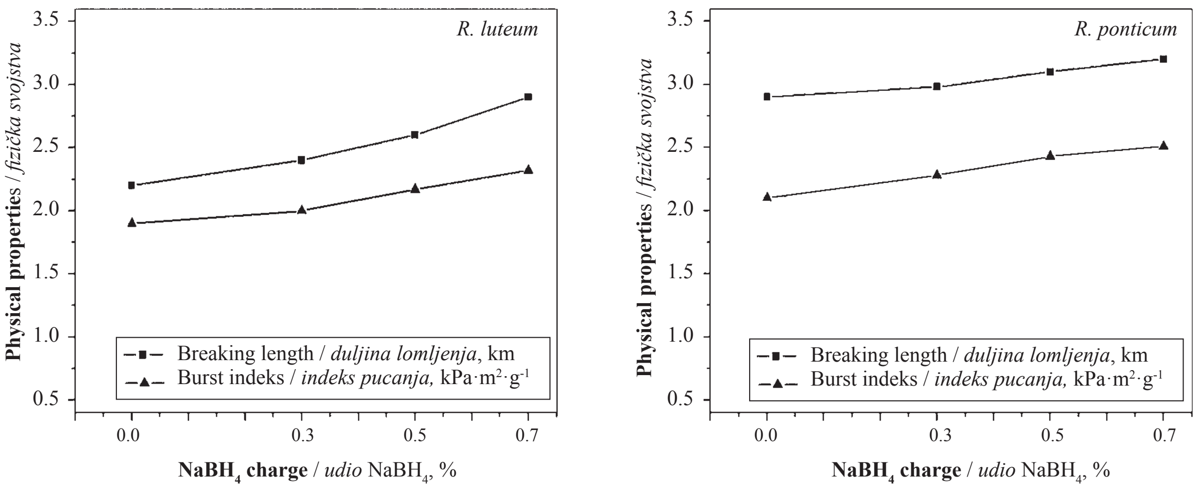

**Figure 2** Effects of NaBH4 charge on breaking length and burst index of Rhododendron papers
**Slika 2.** Utjecaj udjela NaBH4 na duljinu lomljenja i indeks pucanja papira od *Rhododendrona*

produced (Akgul *et al.*, 2007; Copur and Tozluoglu, 2008; Istek and Ozkan, 2008; Istek and Gonteki, 2009; Gumuskaya *et al.*, 2011; Erisir *et al.*, 2015; Gulsoy *et al.*, 2016). With the addition of 0.5 % NaBH$_4$, the breaking lengths of *R. luteum* and *R. ponticum* pulps increased by approximately 16.1 % and 6.4 %, and burst indices also increased by 14.2 % and 15.7 %, respectively. According to data in Tables 3 and 4, optimum cooking conditions for the two species in NaBH$_4$-added cooking experiments were determined by using 0.5 % NaBH$_4$ and 18 % active alkali (Cooking No: 7).

## 4 CONCLUSIONS
## 4. ZAKLJUČAK

In this study, pulp and papers of *R. luteum* and *R. ponticum* species were produced by using Kraft-NaBH$_4$ method and their properties were examined. As a result of this study, it was determined that the chemical properties of *R. luteum* and *R. ponticum,* such as yield, kappa and viscosity, were found to be better for pulp and paper industry. Besides, when the physical properties of the produced papers are examined, it is seen that they are suitable for the production of many paper types. The short-fiber pulps obtained from *R. luteum* and *R. ponticum* can be mixed with long-fiber pulps in certain proportions and evaluated for the production of many paper types. Since 70 % of short-fiber raw materials are used for the production of writing-printing paper, the possibility of using *R. luteum* and *R. ponticum* species for the production of different types of quality paper is quite high.

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# Some Properties of Eastern Red Cedar (*Juniperus Virginiana* L.) Particleboard Panels Made Using Modified Starch as Binder

## Svojstva iverice izrađene od drva crvenog cedra i modificiranog škroba kao veziva

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**ABSTRACT •** *The objective of this study was to evaluate physical and mechanical properties of experimental panels manufactured from eastern red cedar (Juniperus virginiana L.) using modified starch as binder. Modulus of elasticity (MOE), modulus of rupture (MOR), internal bond strength (IB), hardness and dimensional stability in the form of thickness swelling and water absorption in addition to surface quality of the samples were tested. Panels were manufactured at three density levels, which are 0.60 g/cm*$^3$*, 0.70 g/cm*$^3$ *and 0.80 g/cm*$^3$*. The highest MOE, MOR and IB values for the samples having 0.80 g/cm*$^3$ *density were determined as 2207.16 MPa, 15.17 MPa and 0.87 MPa, respectively. Thickness swelling values of the samples soaked in water for 2 h ranged from 15.38 % to 23.23 %. Micrographs taken on the samples using scanning electron microscope revealed that starch was uniformly distributed within the particles. Based on the findings in this study, it appears that eastern red cedar would have a potential as a raw material to manufacture particleboard panel using modified starch as green adhesive with accepted physical and mechanical properties.*

***Keywords:*** *particleboard; green adhesive; starch; mechanical properties; physical properties*

**SAŽETAK •** *Cilj istraživanja bio je odrediti fizička i mehanička svojstva eksperimentalnih ploča proizvedenih od drva crvenog cedra (Juniperus virginiana L.) uz upotrebu modificiranog škroba kao veziva. U eksperimentu su na osnovi debljinskog bubrenja i upijanja vode ispitivani modul elastičnosti (MOE), modul loma (MOR), međuslojna čvrstoća (IB), tvrdoća i dimenzijska stabilnost, kao i kvaliteta površine uzoraka. Ploče su proizvedene u tri gustoće: 0,6; 0,7 i 0,8 g/cm*$^3$*. Najveće vrijednosti MOE, MOR i IB za uzorke gustoće 0,8 g/cm*$^3$ *bili su redom: 2207,16 MPa; 15,7 MPa i 0,87 MPa. Vrijednosti debljinskog bubrenja uzoraka potapanih u vodi tijekom dva sata kretale su se u rasponu od 15,38 do 23,23 %. Mikrografije snimljene na uzorcima elektronskim pretražnim mikroskopom otkrile su da je škrob ravnomjerno raspoređen unutar iverja. Na temelju rezultata ovoga istraživanja, čini se da bi drvo crvenog cedra moglo biti potencijalna sirovina za proizvodnju iverica, uz uporabu modificiranog škroba kao ekološki prihvatljivog ljepila poželjnih fizičkih i mehaničkih svojstava.*

***Ključne riječi:*** *iverica; ekološki prihvatljivo ljepilo; škrob; mehanička svojstva; fizička svojstva*

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## 1 INTRODUCTION
## 1. UVOD

Wood composite panels, such as particleboard, fiberboard, and oriented strand board, are widely used for both non-structural and structural applications. Particleboard is the oldest composite panel that still has a significant market share among the other types of panels. It is mainly used for furniture, cabinet and paneling manufacture in addition to substrate for thin overlays. Most of the time, small diameter logs of softwood and hardwood species, such as pine, aspen or waste from sawmills, are used as raw material for particleboard production. Formaldehyde based adhesives, including urea formaldehyde (UF), are the most used binder in particleboard manufacture due to their low cost and excellent adhesion performance. On the other hand, interior panels, namely particleboard and fiberboard with different density, are two prime products manufactured using UF, which have been on commercial market not only in the USA but also in Europe and Asia for decades. Although UF is the most used adhesive in composite panel production, having formaldehyde in its chemical structure creates important environmental and health issues, since formaldehyde is a possible cancerogenic substance.

Most effective way to omit formaldehyde emission from wood based panels is to use formaldehyde-free or bio-based resins like starch-based resins. Starch is a carbohydrate material that consists of amylase and amylopectin, which could be differentiated by its chemical structure. The linear a-(1 → 4) linked glucan is called amylase, while an a-(1→ 4) linked glucan with 4.2-5.9 % a-(1 → 6) branch linkages is amylopectin (Robyt, 2008; Amini *et al.*, 2013). It can be obtained from various plant materials, such as corn, potato, rice, wheat, sago and many others, and it is widely available throughout the world as it is commonly used in food industry. Therefore, modification of starch was well documented by many researchers (Verwimp *et al.*, 2004; Amini *et al.*, 2013; Yu *et al.*, 2013).

Chemical modification allows starches to be green adhesive solvent in furniture industry. Esterification, etherification, and cross-linking are well-known reactions for chemical modification of starch. These includ succinylation (Phillips *et al.,* 2000), maleination (Chong *et al.,* 2001), and acetylation (Phillips *et al.,* 1999), which can enhance starch functional value and broaden its range of physico–chemical properties. Chemical modification of starch is commonly carried out in aqueous media, water being considered a green, environmentally friendly solvent (Bodîrlău *et. al.,* 2014; H`ng *et al*., 2011).

The chemical modification reactions determine the cross-linking process, which is a common approach to improve the performance of starch for various applications (Huang *et al.,* 2007; Wang *et al.,* 2010). Determination of the modification degree (Phillips *et al.,* 2000) and classification of chemically modified starches using infrared spectros-copy has been described (Dolmatova *et al.,* 1998; Dupuy *et al.,* 1997).

Eastern red cedar (*Juniperus virginiana* L.) is considered an invasive species encroaching in the Great Plains and one of the predominant woody species in the United States. It is also one of widely distributed species in Oklahoma. Current area covered by Eastern red cedar in Oklahoma is estimated around 4.5 million hectares and it is projected to be more than 6.3 million hectares within next 10 years. It is claimed that the red cedar has a growing rate of 380 hectares per day resulting in a significant adverse impact on ecology (Ulker and Hiziroglu, 2017). Generally, red cedar trees are relatively low value trees due to their irregular growth pattern; therefore, lumber manufacture from eastern red cedar is limited to trees with larger diameter. However, red cedar trees can still be used to manufacture different products like indoor and outdoor furniture units, paneling, novelty items, mulch and even composite panels. Different types of experimental particleboard, waferboard and sandwich type of panels, were produced in previous studies (McKinley and Blair, 2008; Reddin and Krementz, 2016; Yang *et al*., 2016; Bragg and Hulbert, 1976; Hiziroglu, 2012; Ulker and Burdurlu, 2016; Hiziroglu *et al*., 2002).

It was determined that both physical and mechanical properties of red cedar particleboard panel products were comparable to those manufactured from different species at commercial scale. Currently, there is very limited or no information on properties of particleboard made from eastern red cedar bonded with modified starch. Therefore, the main objective of this work was to determine the basic properties of the particleboard made from eastern red cedar bonded with green adhesive.

## 2 MATERIALS AND METHODS
## 2. MATERIJALI I METODE

Commercially produced eastern red cedar (*Juniperus virginiana* L.) particles supplied by a local producer in Oklahoma were used to produce experimental panels. Dried particles having a moisture content of 3 % were classified into fine and coarse sizes on a 0.841 mm (20 mesh) and 0.250 mm (60 mesh) screen, respectively. Coarse particles were used for the core layer of the three-layer particleboard, while the fine particles were used for the surface layers of the panel.

Glutardialdehyde (GDA), which was used as starch modifier in our study, is a colorless oily liquid organic compound having the formula $CH_2(CH_2CHO)_2$. GDA is widely used as a disinfector agent for medical equipment. Glutardialdehyde (GDA) acts as a curing agent and plays an important role in uniform distribution of starch on the particles. GDA used for modification of corn starch, was mixed and stirred at room temperature. Corn starch powder was dissolved in distilled water (100 ml) at a temperature of 30 ºC before it was mixed with GDA. The share of the added GDA was 25 % based on volume. No wax or any other additives were used in the panel production.

The ratio of the surface layer mass to the total mass of a panel, known as the shelling ratio, was 0.25 for all sample panels. Shelling ratio was determined

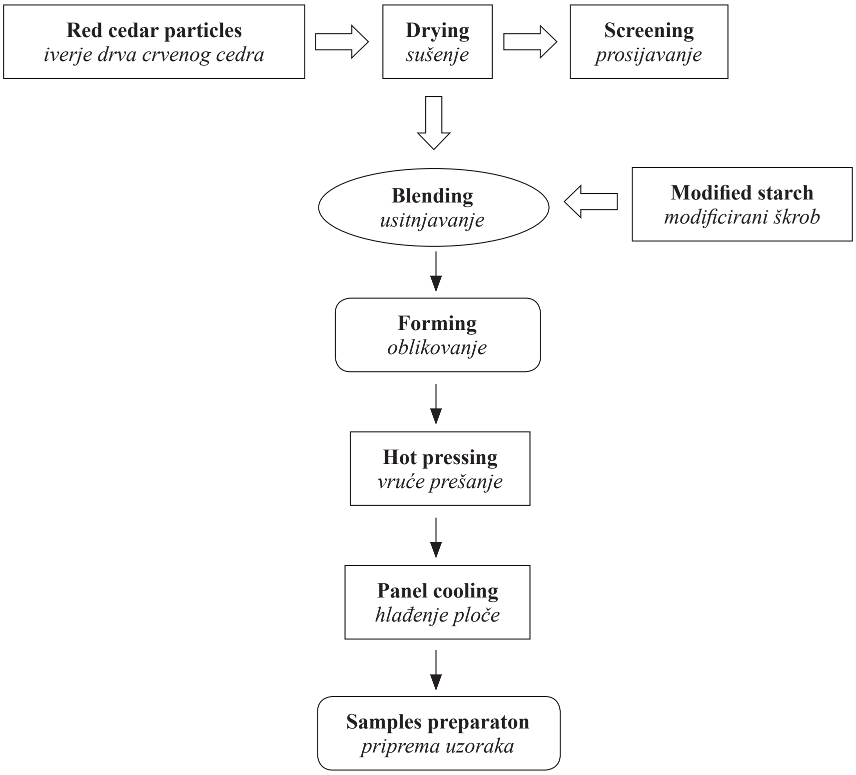


**Figure 1** Panel manufacturing process
**Slika 1.** Proces proizvodnje ploče

using precision scales. Hand formed mats were pressed in a computer-controlled press (Erie Mill and Press Company, Inc., 60.9 cm by 60.9 cm 270-ton press) using a pressure of 5.5 MPa at a temperature of 180 °C, for 5 minutes. A total of fifteen panels were made for each of the three density levels, which were 0.60 g/cm$^3$, 0.70 g/cm$^3$ and 0.80 g/cm$^3$. The process of panel manufacturing is presented in Figure 1. Panels were conditioned in a climate room at 65 % relative humidity and a temperature of 20 °C for two weeks before the samples were cut for different tests based on ASTM (American Society for Testing Materials) D1037-12 (ASTM International, 2012).

The mechanical properties of the specimens were carried out on a Comten Testing System, Model 95-VD equipped with 1000 kg load cell. ASTM D1037-12 procedures were used to determine bending strength and internal bond strength test. The samples with the size of 50 mm by 300 mm by 12 mm from each panel type were tested for their bending strength properties.

Internal bond strength (*IB*) of the panels was determined using sample 50 mm by 50 mm cross sections. Hardness of the sample panels was tested by embedding a hemisphere steel having 11.2 mm diameter onto surface of panels using Comten 95 Series Universal Testing machine. Surface characteristics of the samples with 50 mm by 300 mm by 12 mm were evaluated by employing a fine stylus profilometer Hommel T-500 unit, which has TkE model pick-up, equipped with a skid type diamond stylus having 5 µm tip radius and a 90° tip angle (Hiziroglu *et al*., 2004). The stylus traversed the surface at a constant speed of 1 mm/s over a 15.2 mm tracing length and the vertical displacement of the stylus was converted into an electrical signal. Average roughness, ($R_a$), mean peak-to-valley height ($R_z$), and maximum peak height value ($R_{max}$) were used for the evaluation of the surface roughness of specimens.

The samples were also examined using a scanning electron microscope, FEI Quanta 600 FEG scanning electron microscope with an EVEX X-ray microanalysis system. The specimens of 5 mm by 10 mm by 6 mm were prepared from the panels of 0.8 g/cm$^3$ density. The images were taken with a SEM operated at 15 kV. All the specimens were coated with thin gold layer prior to the analysis.

Thickness swelling (*TS*) and water absorption (*WA*) tests were carried out by soaking specimens of 100 mm by 100 mm by 12 mm in distilled water for 2 and 24 h based on ASTM D1037-12 standard. The specimens were submerged horizontally under 25 mm of water maintained at a temperature of (20±1) °C. After a 2 h submersion, the specimens were taken out to drain for (10±2) min, then the excessive water was removed and they were immediately weighed. Their thickness was measured at four corners at accuracy of 0.01 mm at each soaking period. Then the specimens were submerged for an additional period of 22 h and the above weighing and measuring procedures were repeated. WA and TS test were carried out to analyze the dimensional stability of panels.

Analysis of variance (ANOVA) was used to analyze the significant differences among the specimens manufactured with three different densities. The data were statistically analyzed at a confidence level of *p*-value = 0.05 and Duncan's multiple range test was also carried out using SAS 9.4 software (SAS Institute Inc., Cary, NC) for multiple comparison.

## 3 RESULTS AND DISCUSSION
## 3. REZULTATI I RASPRAVA

Average and standard deviation values (in parentheses) of mechanical and physical properties of the samples are presented in Table 1.

Panel type A with the density of 0.80 g/cm$^3$ resulted in the highest *MOE*, *MOR* and IB values of 2207.16 MPa, 15.17 MPa and 0.87 MPa, respectively. These values were followed by panel type B and C as a function of their density levels. It is a well-known fact that the overall density influences most the mechanical properties of particleboards (Kelly, 1977; Ulker and Burdurlu, 2016; Ayrilmis, 2007; Cai and Ross, 2010). Both bending and *IB* strength values of experimental samples increased with increasing the density levels of the panels. Overall findings in this work also confirmed such trend. In a previous work, carried out using eastern red cedar as raw material and a combination of urea formaldehyde and modified starch as binder in particleboard manufacture, *MOE* and *MOR* values were 2344.32 MPa and 12.14 MPa, respectively (Chotikhun and Hiziroglu, 2017).

Experimental panels with the density of 0.80 g/cm$^3$ of the same type of raw material mixed with 7 % urea formaldehyde resulted in 2.31 MPa for the above values. It seems that using only modified starch as a binder in the panels adversely influences their bending characteristics. Table 2 displays statistical analysis of *MOE*, *MOR*, and *IB* values of the samples.

Based on ANOVA, it was found that significant difference existed among the properties of the samples as a function of their density levels. Table 3 gives the means for groups of homogeneous subsets.

As mentioned above, mechanical properties of the panels with the density of 0.80 g/cm$^3$ and 0.70 g/cm$^3$ were found satisfactory based on TS-EN 312 (Particleboards Specifications) standard, while the particleboard with the density of 0.60 g/cm$^3$ does not meet the requirements of TS-EN 312 standard.

Dimensional stability in terms of thickness swelling and water absorption of the samples resulted in relatively low values. Thickness swelling of the samples ranged from 15.38 % to 31.40 % as a result of 2 and 24 h water soaking. The lowest thickness swelling of 15.38 % was determined for the panels with the density of 0.8 g/cm$^3$ exposed to 2 h water soaking. The highest thickness swelling of 31.40 % was found for the panels with the density of 0.60 g/cm$^3$ at 24 h water soaking. In a previous work, thickness swelling values of particleboard panels, made using a combination of starch and 2 % UF, ranged from 26 % to 37 % (Chotikhun and Hiziroglu, 2017; Garay, 2012). Water absorption values of the samples ranged from 116.80 % to 164.30 % as a function of 2 and 24 h water soaking. Past studies that used different types of raw materials and starch as a binder also found comparable thickness swelling and water absorption values as those found in this study (Chotikhun and Hiziroglu, 2017). It is a known fact that any kind of starch, including corn

**Table 1** Average values of mechanical and physical properties of samples (Standard deviation given in parentheses)
**Tablica 1.** Srednje vrijednosti mehaničkih i fizičkih svojstava uzoraka (u zagradama su standardne devijacije)

| **Panel type** *Vrsta ploče* | **Density g/cm$^3$** *Gustoća* g/cm$^3$ | **Bending, MPa** *Savijanje,* MPa | | **IB strength, N/mm$^2$** *Međuslojna čvrstoća,* N/mm$^2$ | **Janka hardness N/mm$^2$** *Tvrdoća po Janki,* N/mm$^2$ | **Dimensional stability** *Dimenzijska stabilnost* | | | | **Surface roughness,** µm *Hrapavost površine,* µm | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | ***MOE* N/mm$^2$** | ***MOR* N/mm$^2$** | | | **Thickness swelling, %** *Debljinsko bubrenje,* % | | **Water absorption, %** *Upijanje vode,* % | | | | |
| | | | | | | 2-h | 24-h | 2-h | 24-h | $R_a$ | $R_z$ | $R_{max}$ |
| A | 0.80 | 2207.16<br>(23.32) | 15.17<br>(22.28) | 0.87<br>(15.77) | 6.00<br>(15.01) | 15.38<br>(16.40) | 18.32<br>(19.57) | 132.17<br>(2.35) | 164.30<br>(3.84) | 11.74<br>(3.03) | 28.49<br>(8.88) | 43.32<br>(13.09) |
| B | 0.70 | 1732.31<br>(15.05) | 13.05<br>(14.29) | 0.78<br>(25.94) | 5.70<br>(16.70) | 15.45<br>(15.23) | 23.10<br>(19.15) | 123.90<br>(1.91) | 160.60<br>(2.94) | 14.30<br>(2.24) | 30.50<br>(10.47) | 48.17<br>(10.38) |
| C | 0.60 | 1136,23<br>(10.92) | 6.18<br>(11.18) | 0.45<br>(12.53) | 5.46<br>(13.55) | 23.23<br>(17.27) | 31.40<br>(19.39) | 116.80<br>(1.65) | 153.70<br>(2.19) | 18.07<br>(3.53) | 37.56<br>(10.70) | 60.54<br>(17.46) |

**Table 2** Statistical analysis of *MOE*, *MOR*, and *IB* values of the samples based on ANOVA
**Tablica 2.** Statistička analiza ANOVA za vrijednosti *MOE*, *MOR* i *IB* uzoraka

| | | **Sum of Squares** *Zbroj kvadrata* | ***df*** | **Mean Square** *Kvadrat srednje vrijednosti* | ***F*** | **Significance** *Značajnost* |
|---|---|---|---|---|---|---|
| *MOE* | Between Groups / *između skupina* | 438978.156 | 2 | 219489.07 | 665.85 | .000 |
| | Within Groups / *unutar skupina* | 8900.211 | 27 | 329.63 | | |
| | Total / *ukupno* | 447878.367 | 29 | | | |
| *MOR* | Between Groups / *između skupina* | 9560381.267 | 2 | 4780190.63 | 15646.91 | .000 |
| | Within Groups / *unutar skupina* | 8248.600 | 27 | 305.50 | | |
| | Total / *ukupno* | 9568629.867 | 29 | | | |
| *IB* | Between Groups / *između skupina* | 21475.616 | 2 | 10737.80 | 26.87 | .000 |
| | Within Groups / *unutar skupina* | 10789.387 | 27 | 399.60 | | |
| | Total / *ukupno* | 32265.003 | 29 | | | |

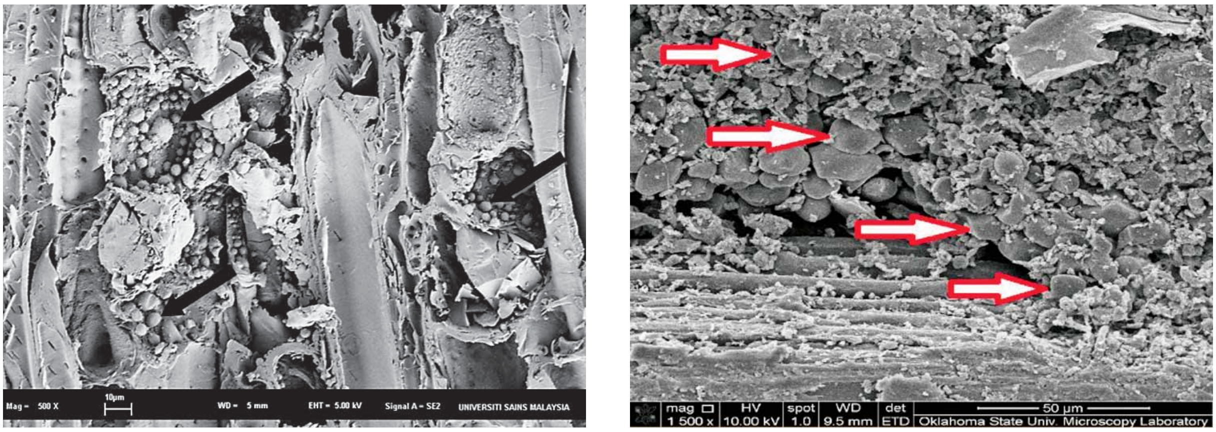
**Table 3** Means for groups of homogeneous subsets
**Tablica 3.** Srednje vrijednosti za grupe homogenih podskupova

| **Mechanical properties**<br>*Mehanička svojstva* | **Panel type**<br>*Vrsta ploče* | **Number of samples**<br>*Broj uzoraka* | **Subset for alpha 0.05**<br>*Podskup za alfa 0,05* | | |
|---|---|---|---|---|---|
| | | | 1 | 2 | 3 |
| *MOE*, MPa | A (0.80 g/cm$^3$) | 10 | 2207.16 | | |
| | B (0.70 g/cm$^3$) | 10 | | 1732.31 | |
| | C (0.60 g/cm$^3$) | 10 | | | 1136.23 |
| *MOR*, MPa | A (0.80 g/cm$^3$) | 10 | 15.17 | | |
| | B (0.70 g/cm$^3$) | 10 | | 13.05 | |
| | C (0.60 g/cm$^3$) | 10 | | | 6.18 |
| *IB*, MPa | A (0.80 g/cm$^3$) | 10 | 0.87 | | |
| | B (0.70 g/cm$^3$) | 10 | 0.78 | | |
| | C (0.60 g/cm$^3$) | 10 | | 0.45 | |

starch used in this work, is hygroscopic material influencing adversely overall dimensional stability of the panels. Additionally, not using any wax in the panels manufactured would be considered as one of the reasons for slightly high thickness swelling and water absorption characteristics of the samples. Although the samples had somehow high dimensional instability, none of them disintegrated or crumbled as a result of water soaking test.

Since most of the particleboards are used as substrate for thin overlays, their surface quality in terms of roughness plays an important role during their service life. The average roughness ($R_a$) value of 11.74 μm was determined for the samples with 0.80 g/cm$^3$. Boards with lower density had rougher surface, which can be related to their less densified surface. A typical commercially manufactured particleboard could have $R_a$ values ranging from 3.80 μm to 5.90 μm (Hiziroglu, 1996; Chotikhun and Hiziroglu, 2017). Also most of the commercially manufactured particleboard panels are sanded with up to 180 grit sand paper, which gives a very uniform smooth surface. None of the samples was sanded in this work. If the surface of the experimental panels had been sanded, surely $R_a$ values, along with other two roughness parametres, would have been much lower.

Amini *et al*. (2013) worked on properties of particleboard made from rubberwood using modified starch as binder, in their publication. Figure 2.1 shows, with black arrows, SEM cross sectional view of particleboard made using glutardialdehyde modified corn starch as a binder at 500 magnification with the presence of modified corn starch granules. In our study, we also had SEM cross sectional view of particleboard made using glutardialdehyde modified corn starch as a binder at 1500 magnification with the presence of modified corn starch granules. Uniform distiribution of starch throughout the particle could be considered as one of the responsible parameters. As can be seen in Figure 2.2, starch was distrubuted quite uniformly among the particles even though manual mixture was used during the panel manufacture.



a) b)

**Figure 2** SEM cross sectional view of particleboard made using GDA modified corn starch as a binder: a) SEM cross sectional view of particleboard made using GDA modified corn starch as a binder at 500 magnification with the presence of modified corn starch granules shown with black arrows (Amini et al., 2013); b) SEM cross sectional view of particleboard made using GDA modified corn starch as a binder at 1500 magnification with the presence of modified corn starch granules shown with white-red colored arrows

**Slika 2.** SEM presjeci iverice proizvedene upotrebom GDA modificiranoga kukuruznog škroba kao veziva: a) SEM presjek iverice proizvedene uz dodatak GDA modificiranoga kukuruznog škroba kao veziva pri povećanju od 500 puta, s vidljivim granulama kukuruznog škroba označenima crnim strelicama (Amini *et al*., 2013.), b) SEM presjek iverice proizvedene primjenom GDA modificiranoga kukuruznog škroba kao veziva pri povećanju od 1500 puta, s vidljivim granulama kukuruznog škroba označenima bijelo-crvenim strelicama

It is a known fact that overall strength properties of particleboard is a function of glueline between the particles. As can be observed from the micrographs taken on SEM, starch was distributed relatively uniformly, which resulted in enhanced bonding among the particles. Such finding was also reflected in overall properties tested in this work.

## 4 CONCLUSIONS
## 4. ZAKLJUČAK

Fundamental mechanical and physical properties of the experimental panels made in this work showed satisfactory values with the exception, to a certain extent, of dimesional stability. It seems that 100 % modified starch with GDA would have a potentional to be used as green adhesive in the manufacture of particleboard. This approach, combined with an invasive species, such as eastern red cedar, would result in green panels that would be particularly attractive in terms of sustainability and environmental friendliness.

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# Effect of Natural Resin from Wild Pistachio Trees on Physical Properties and Durability of Beech Wood: Alone and in Combination with Boric Acid

## Utjecaj prirodne smole divljeg drva pistacije (same i u kombinaciji s bornom kiselinom) na fizička svojstva i trajnost bukovine

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**ABSTRACT •** *This study was carried out to investigate the physical properties and decay resistance of beech wood treated with natural pistachio resin (PR) from Iranian wild pistachio trees (Pistacia atlantica), alone and in combination with boric acid (BA). Wood samples were impregnated with different concentration of PR dissolved in ethanol (3 to 20 %) with vacuum-pressure technology. The combination of PR (20 %) and BA (2 %) was also conducted to evaluate any interaction or synergistic effects. The water absorption, volumetric swelling, and decay resistance against Trametes versicolor fungi, before and after a leaching test (EN 84), were measured on treated and untreated samples. The chemical compositions of PR were also identified by gas chromatography–mass spectrometry (GC-MS) techniques. The chemicals analysis identified more than 20 different compounds in the PR, monoterpenoids being the predominant fraction and α-pinene the major component. The samples treated with a higher concentration of PR showed much higher weight gain percentage (WG%). The results showed that the increase in WG% reduced the average values of water absorption and volumetric swelling of treated samples even after long terms of soaking in water. The decay resistance of the treated samples increased against white rotting fungi as the values of WG% increased. Efficient protection was seen when a combined treatment of PR and BA was used. Even after the leaching process, the weight loss of the treated samples was less than 3 percent. The samples treated with BA alone largely lost their effectiveness against fungal attack after the leaching. The use of PR along with an environmental friendly co-biocide can also be recommended for wood preservation in places that require minimal toxicity.*

***Keywords:*** *pistachio trees; natural resin; weight gain; durability; physical properties*

**SAŽETAK •** *Cilj ovog rada bio je istražiti fizička svojstva i otpornost na propadanje bukovine tretirane prirodnom smolom pistacije (PR) dobivene iz divljeg drva pistacije koja uspijeva u Iranu (Pistacia atlantica) te kombinaci-*

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*jom smole pistacije i borne kiseline (BA). Uzorci drva vakuumsko-tlačnim su postupkom impregnirani različitim koncentracijama PR-a otopljenoga u etanolu (3 do 20 %). Provedena je i impregnacija uzorka kombinacijom PR-a (20 %) i BA-a (2 %) kako bi se procijenili svi interakcijski i sinergijski učinci. Za tretirane i netretirane uzorke mjerena je upojnost vode, volumetrijsko bubrenje i otpornost na djelovanje gljive Trametes versicolor prije i nakon ispiranja (EN 84). Ujedno je uz pomoć plinske kromatografije s masenom spektrometrijom (GC-MS) utvrđen i kemijski sastav smole (PR). Kemijskom analizom u PR-u je identificirano više od 20 različitih spojeva; momoterpeni su bili dominantna frakcija, a* a*-pinen glavna komponenta. Uzorci tretirani većom koncentracijom PR-a rezultirali su većim dobitkom mase (WG%). Rezultati su pokazali da se s porastom WG% smanjuje prosječna vrijednost upojnosti vode i volumetrijsko bubrenje tretiranih uzoraka, čak i nakon dugotrajnog potapanja u vodi. Otpornost tretiranih uzoraka na djelovanje gljiva bijele truleži povećala se s povećanjem WG%. Učinkovita zaštita primijećena je pri primjeni kombiniranog tretmana PR-om i BA-om; čak je i nakon postupka ispiranja gubitak mase tretiranih uzoraka bio manji od 3 %. Uzorci tretirani samo BA-om nakon ispiranja su uglavnom izgubili otpornost na napad gljiva. Moguće je zaključiti da se za zaštitu drva na mjestima koja zahtijevaju minimalnu otrovnost može preporučiti upotreba PR-a zajedno s ekološki prihvatljivim biocidom.*

***Ključne riječi:*** *drvo pistacije; prirodna smola; dobitak mase; trajnost; fizička svojstva*

## 1 INTRODUCTION
## 1. UVOD

While wood is a unique biomaterial that currently has many uses, it also has some disadvantages that limit its use for some applications. Some wood species are prone to be readily degraded by fungi, termites, and other organisms (Reinprecht, 2016). Wood protection is a process for reducing and/or preventing attack by wood deteriorating agents to increase its service life (Jones and Brischke, 2017). CCA (chromium (VI)-copper-arsenic) formulations have been widely used for the treatment of wood (Freeman *et al*., 2003). However, CCA-treated lumber contains arsenic, which may pose serious health risks even after its service time ends and the CCA treated wood becomes waste. As a result, arsenic (and of course CCA) has been banned since 2004 from most applications for wood preservation in Europe and North America (Caldeira, 2010). Environmental performance and sensitivity play an increasing role in the development and use of wood preservatives (Shmulsky and Jones, 2011). There are several arsenic-free alternatives for CCA, and although their main ingredients have lower mammalian toxicity than arsenic, these systems contain high levels of copper, which can be toxic to aquatic life (Lebow, 2004). Therefore, many studies are underway to replace metal oxides with more eco-friendly biocides.

In recent years, many studies have been conducted on the use of plant extracts for wood preservation. Singh and Singh (2012) have reviewed many of these studies, and concluded that progress in implementation of the technologies has been slow because of certain limitations. It was found that the essential oil from *Lippia origanoides* showed high fungicidal activity against two wood decay fungi. This activity was attributed to thymol, the major component of the essential oil. The U.S. Environment Protection Agency (EPA) has no significant incident reports involving thymol (De Medeiros *et al*., 2016). In another study, 16 plant essential oils were applied to *Fagus orientalis* and *Pinus taeda* wood samples by vacuum impregnation method (Bahmani and Schmidt, 2018). The treated samples were then infected with different kinds of rotting and molding fungus. Their results showed that lavender oil, lemon grass oil, and thyme oil had the greatest effect against mold and wood decay fungi. Yang and Clausen (2007) evaluated the ability of seven essential oils to inhibit the growth of some molding fungi on southern yellow pine that were either dip treated or exposed to vapors of the test oils. Thyme and Egyptian geranium oil inhibited the growth of all test fungi. The extract of cinnamon leaves has also proven to be highly effective against wood decay fungi and termites (Cheng *et al*., 2006). It should be noted, however, that breathing, eating, or touching the cinnamon products can trigger an allergic reaction in some people. The effect of cinnamon, clove, anise, lime, and tangerine oils on the control of mold fungi were also studied on rubber wood (Matan and Matan, 2007, 2008).

The impact of plant extracts and essential oils on wood growing molds have been extensively studied in recent years (Salem *et al*., 2016). Virtually none of the oils have been commercialized, but they may be used especially for artistic or domestic uses.

A number of studies have also suggested that a combination of plant extracts with more environmentally friendly preservatives may achieve positive interactions or synergy effects. The modification of mimosa tannin with a copper-ammonia complex appeared to promote anti-decay properties of treated wood, while unmodified tannin had no preventive effect on fungal attack (Yamaguchi and Okuda, 1998; Yamaguchi and Yoshino, 2001).The association between tannins and boric acid could also be another remedy. Pizzi and Baecker (1996) used preservative solutions based on tannins and boron in which boric acid was used to induce hardening reactions of polyflavonoid tannins. Equally, boric acid can be partially fixed into the wood by being trapped in the tannin polymer (Efhamisisi *et al.*, 2017).

Boron compounds, such as boric acid, have been used for over 40 years as wood preservatives in Australia and Europe (Caldeira, 2010). Borates offer some of the most effective and versatile wood preservative systems available today, combining the properties of broad spectrum efficacy and low mammalian toxicity (Freeman *et al*., 2003). Due to the favorable environmental characteristics of borates, researchers have refocused on boron-including compounds in the last two decades (Obanda *et al*., 2008). However, more recently (August, 2008), the European Commission decided to make an ATP – Adaptation to Technical Progress of Council Directive 67/548/EEC (the 30th ATP), and since then boric acid and sodium borate have been classified as reprotoxic. This means that these borate compounds are now classified as substances toxic for reproduction category 2 for both fertility and developmental effects. According to this restriction, the concentration limit for boric acid is ≥ 5.5 % for wood treatment. However, borates have been registered by the EPA as wood preservatives. Also, they are still in use in Europe, but the concentration is limited. High susceptibility of borates to leach is the main obstacle to their widespread use as a major component in the broad spectrum (Obanda *et al*., 2008).

*Pistacia* is a genus of flowering plants from the *Anacardiaceae* family that can be found in about twenty species (Bozorgi *et al*., 2013). Extracts of *Pistacia atlantica* could be considered as a potential alternative to antifungal chemicals that have a detrimental effect on the environment and public health (Amri *et al*., 2015). On the other hand, it was found that natural materials alone cannot provide full protection.

There are no exact statistics on the removing of resin from wild pistachio trees in Iran. These trees are found in Zagros, the forests located in all the western provinces of the country from north to south. Generally, about 7,000 tons of this resin is produced annually in Iran, with more than seventy percent being exported overseas without any processing (Jahanbazi *et al*., 2006). Zagros forests, with an area of about 6 million hectares (3.5 percent of Iran), are located in the west of the country (Sadeghi *et al*., 2017). Wild pistachio trees are considered the most important species in Zagros forests (Javanmiri Pour *et al*., 2013). In this study, the resin of pistachio trees, grown in the western forests of Iran, was used as wood preservative material. Since the natural materials cannot provide full protection alone, the pistachio resin was used both alone and in combination with boric acid. Although boric acid is easily leached from treated wood (Obanda *et al*., 2008), it seems that its combination with pistachio resin, as a water insoluble material, prevents its leaching.

## 2 MATERIALS AND METHODS
## 2. MATERIJALI I METODE

### 2.1 Materials
### 2.1. Materijali

The natural resin of wild pistachio (*Pistacia atlantica*) trees was obtained from an area located at 45°30′ to 46˚15′ longitude and 35°45′ to 36°15′ altitude in the Kurdistan province, western Iran. The average elevation of the area stands 1400 m above sea level. The average annual precipitation is 300 to 800 mm/year and average annual temperature is between 12 to 18 °C. Boric acid of 99 % purity was purchased from the Sigma Aldrich Company and ethanol of 96 % purity was purchased from the Merck Company. The sapwood of beech (*Fagus orientalis*) was prepared from the Hezarjeribs forests of Mazandaran province in northern Iran. The climate of this region is mild and humid and wood specimens were harvested at the altitude of 1000 m. The chosen specimens were free from cracks, stain, decay, insect damage, and other defects. The samples were then prepared according to relevant standards (Table 1).

### 2.2 Methods
### 2.2. Metode

### 2.2.1 Gas chromatography-mass spectrometry (GC-MS) of the resin
### 2.2.1. Plinska kromatografija s masenom spektrometrijom (GC-MS) za smolu

The ingredients of the resin were determined with a gas chromatography/mass selective detector (GC-MS). The PR was firstly dissolved in ethyl ether before injection into the GC apparatus. The analysis was performed on an Agilent 7890A gas chromatograph. The separation was achieved with a Rtx 5MS capillary column (30 m × 0.25 mm; 0.25 μm film thickness) with He as the carrier gas (1.3 ml/min). The oven temperature was programmed from 40 to 200 °C at 5 °C/min. The temperatures of the injector and quadrupole and ion sources were 250 °C, 150 °C, and 230 °C, respectively. The MS detector was run in electron impact mode with electron energy of 70 eV. Volatile organic compounds were identified by comparison of their retention indices, calculated by the use of a series of n-alkanes (C9–C16), with those reported in the literature.

### 2.2.2 Sample preparation
### 2.2.2. Priprema uzoraka

Different treatment solutions were prepared by solving PR in ethanol with different concentrations of 3, 6, 9, 12, 15 and 20 %. One treatment solution contained 20 % PR and 2 % boric acid, and the other contained 2 % boric acid without any PR. The components of the treatment solutions and their nomenclature are shown in Table 2. Specimens were treated with the vacuum-pressure method (Dhamodaran and Gnanaharan, 2007). The samples were put inside the cylinder under 0.8 bar vacuums for 30 minutes and then 4 bar pressure was applied for 2 hours at 50 °C. Afterward, the treated samples were kept at lab ambience for 2 weeks to evaporate the solvent and then were oven dried at (103±2) °C.

**Table 1** Standards used in different tests
**Tablica 1.** Standardi primijenjeni za različita ispitivanja

| **Test**<br>*Ispitivanje* | **Number of treatment**<br>*Broj tretmana* | **Dimensions (L×R×T), mm**<br>*Dimenzije (L×R×T)*, mm | **Standard** |
|---|---|---|---|
| Fungal attack test / *napada gljiva* | 12 | 15×25×50 | EN 113 (1996) |
| Physical test / *fizičkih svojstava* | 10 | 20×20×20 | ISO 13061 (2014) |

**Table 2** Components of treatment solution and their nomenclature
**Tablica 2.** Komponente u otopini za tretiranje i njihova nomenklatura

| **Samples ID**<br>*Oznaka uzorka* | **Concentration in solution**<br>*Koncentracija u otopini*<br>wt% | |
|---|---|---|
| | Resin<br>*Smola* | Boric acid<br>*Borna kiselina* |
| Control (Solvent treated)<br>*kontrolni uzorak (tretirano otapalom)* | 0 | 0 |
| PR3 | 3 | 0 |
| PR6 | 6 | 0 |
| PR9 | 9 | 0 |
| PR12 | 12 | 0 |
| PR15 | 15 | 0 |
| PR20 | 20 | 0 |
| PR20BA2 | 20 | 2 |
| BA2 | 0 | 2 |

### 2.2.3 Measurement of weight gain and retention
### 2.2.3. Mjerenje dobitka mase i retencije

Since some extractive content of wood is removed from the samples during the impregnation process, which causes a mistake in the calculation of *WG*, *WG* values were calculated by two methods. The first was the theoretical weight gain ($WG_t$), which was calculated based on the wet weight after impregnation and concentration of materials in the solutions (Eq.n 1). The second was the actual or experimental weight gain ($WG_e$), which was based on the oven dried samples after impregnation (Eq. 2).

$$WG_t(\%) = \frac{(M_1 - M_0)\cdot C}{M_0}\cdot 100 \tag{1}$$

Where, $M_1$ is the wet weight of the sample after impregnation (g); $M_0$ is the oven dried weight before impregnation (g); and C is the solid percent content in solvent (%).

$$WG_e(\%) = \frac{M_2 - M_0}{M_0}\cdot 100 \tag{2}$$

Where, $M_2$ is the oven dried weight after impregnation (g) and $M_0$ is the oven dried weight before impregnation (g).

Also, the retention ($R_t$) of materials as kilogram per cubic meter of treated wood sample was calculated based on Eq. 3:

$$R_t\left(\text{kg}\cdot\text{m}^{-3}\right) = \frac{(M_2 - M_0)\cdot C}{V}\cdot 10^{-3} \tag{3}$$

Where, $M_2$ is the oven dried weight of the sample after impregnation (g); $M_0$ is the oven dried weight before impregnation (g); $C$ is the solid percent content in solvent (%); and $V$ is the volume of the sample ($m^3$).

### 2.2.4 Water absorption and volumetric swelling measurements
### 2.2.4. Mjerenje upojnosti vode i volumetrijskog bubrenja

The treated and untreated specimens were placed in beakers, which were then filled with distilled water and maintained at 23 °C for 8 days. During that time, the water was replaced by fresh distilled water at 2, 4, 6, 8, and 24 h and then replaced again at 24 h intervals. The weight and volume of the samples were measured at the end of the treatment. The water absorption (*WA*) of the samples was calculated according to Eq. 4.

$$WA(\%) = \frac{(W_1 - W_0)}{W_0}\cdot 100 \tag{4}$$

Where, $W_1$ is the weight of the specimen after immersion in water; $W_0$ is the oven dried weight before immersing in water.

The volumetric swelling (*S*) of the samples was calculated according to Eq. 5.

$$S(\%) = \frac{(V_1 - V_0)}{V_0}\cdot 100 \tag{5}$$

Where, $V_1$ is the volume of the sample after immersion in water and $V_0$ is the volume of the sample before immersion in water.

### 2.2.5 Leaching test
### 2.2.5. Test ispiranja

A leaching test for treated wood was carried out according to the EN 84 standard. In this way, deionized water fivefold the sample volume was added to the samples and exposed to 4 kPa vacuums for 20 minutes. The water was replaced with fresh water after two hours and thereafter at 24 h intervals for 14 days. Weight loss of the samples were then measured.

### 2.2.6 Wood decay test
### 2.2.6. Ispitivanje propadanja drva

The treated and untreated specimens were tested for resistance to biological attack according to the European EN 113 standard criteria, 1996, against *Trametes versicolor* (tropical white rot) grown on malt-agar medium. The *Trametes versicolor* fungi, strain CTB 863A were prepared from Tehran university fungi bank. The wood samples were dried at 105 °C for 24 h and the dry weight of sample was measured before decay testing. Petri dishes (145 mm diameter) with malt extract agar (50 ml per plate) were inoculated with a mycelium agar disc (5 mm diameter) taken from the sub-margin of 1-month-old cultures of *T. versicolor*. When the fungal mycelium reached the border of the plate, three untreated and three treated wood samples were added on separate metal grids to prevent moisture uptake by the wood sample. The plates were incubated at (22±2) °C and (70±5) % relative humidity for 16 weeks. Twelve wood samples for each treatment from four different plates were used to determine the mass loss of the wood samples. After incubation, the sample surfaces were cleaned gently and dried at 105 °C for 24 h to obtain the dry weight of sample after decay testing. The weight loss (*WL*) of the samples was calculated by Eq. 6.

$$WL(\%) = \frac{T_0 - T_1}{T_0}\cdot 100 \tag{6}$$

Where, $T_0$ is the oven dried weight of sample before fungi test (g) and $T_1$ is the oven dried weight after fungi test (g).

## 3 RESULTS AND DISCUSSION
## 3. REZULTATI I RASPRAVA

### 3.1 Pistachio resin (PR) analysis
### 3.1. Analiza smole iz pistacije (PR)

The GC-MS analysis of the PR identified more than 20 compounds. Total number of components and their retention times are shown in Table 3 and Figure 1.

In their study of *Pistacia atlantica* resin from the Kordestan province in Iran, Mahjoub *et al.* (2018) reported 82.64 % for α–pinene as the main constituent and 3.04 % for β-pinene, while Barrero *et al.* (2005) obtained 42.9 % for α–pinene and 13.2 % for β-pinene as the major components of Moroccan *Pistacia atlantica* oil. Benabderrahmane *et al.* (2016) reported 79.8, 65.3, and 25.4 % for α–pinene content of *pistacia atlantica* growing wild in three different regions in the west and north-west of Algeria.

Comparison with previously studied essential oil of the resin from different species of *Pistacia* showed that monoterpenes are the predominate fraction and α-pinene is the major component.

### 3.2 Weight gain and retention analyses
### 3.2. Analiza dobitka mase i retencije

*WG*$_t$ and *WG*$_e$ of the treated samples are presented in Figure 2. As can be seen, the values of *WG* increased as the PR concentration increased from 3 to 20 %. The negative values of *WG*$_e$ for the solvent treated samples may be due to the removal of wood extractives by the ethanol during the impregnation process.

The values of *R*$_t$ increased from 11.86 to 78.34 kg/m$^3$ by increasing resin concentration from 3 to 20 %, respectively (Figure 3).

Lesar *et al.* (2009) indicated that boric acid retention of 0.4 kg/m$^3$ was enough to inhibit the growth of *A. vaillantii*, *S. lacrymans,* and *T. versicolor,* while the retention of 0.8 kg/m$^3$ of boric acid was needed to preserve wood against *P. ostreatus* and *H. fragiforme*

**Table 3** Components of PR
**Tablica 3.** Komponente u PR-u

| **Compounds**<br>*Spojevi* | **Retention time**<br>*Vrijeme retencije*<br>min | **Area**<br>*Površina*<br>% |
|---|---|---|
| meta-Xylene | 6.27 | 0.52 |
| (-)-alpha-Pinene | 7.975 | 85.61 |
| Camphene | 8.397 | 0.70 |
| Bicyclo[2.1.1]hex-2-ene, 2-ethenyl | 8.574 | 0.23 |
| beta-Pinene | 9.216 | 1.72 |
| Cyclotetrasiloxane, octamethyl | 9.824 | 0.24 |
| 3,7,7-Trimethylbicyclo[4.1.0] hept-3-ene | 10.204 | 0.58 |
| p-Cimene | 10.651 | 0.36 |
| L-Limonene | 10.761 | 0.69 |
| Eucalyptol | 10.854 | 0.30 |
| alpha-Terpinene | 12.55 | 1.94 |
| Verbenene | 13.141 | 0.37 |
| 1,7,7-Trimethylbicyclo[2.2.1] hept-5-en-2-ol | 13.699 | 0.22 |
| Trans-Pinocarveol | 14.104 | 0.45 |
| Bicyclo[3.1.1]heptan-3-ol, 6,6-dimethyl-2-methylene | 14.273 | 0.57 |
| Cyclopentasiloxane, decamethyl | 14.408 | 0.32 |
| p-Mentha-1,5-dien-8-ol | 14.931 | 0.45 |
| 4-Terpineol | 15.21 | 0.19 |
| Benzenemethanol, alpha,alpha,4-trimethyl | 15.488 | 0.42 |
| Alpha terpinolene | 15.606 | 1.32 |
| Myrtenol | 15.775 | 0.49 |
| Bicyclo[3.1.1]hept-2-en-6-one, 2,7,7-trimethyl | 16.155 | 0.61 |
| Isobornyl acetate | 18.223 | 0.78 |
| Nonadecane | 28.117 | 0.42 |
| Tetracosane, 2,6,10,15,19,23-hexamethyl | 28.227 | 0.45 |

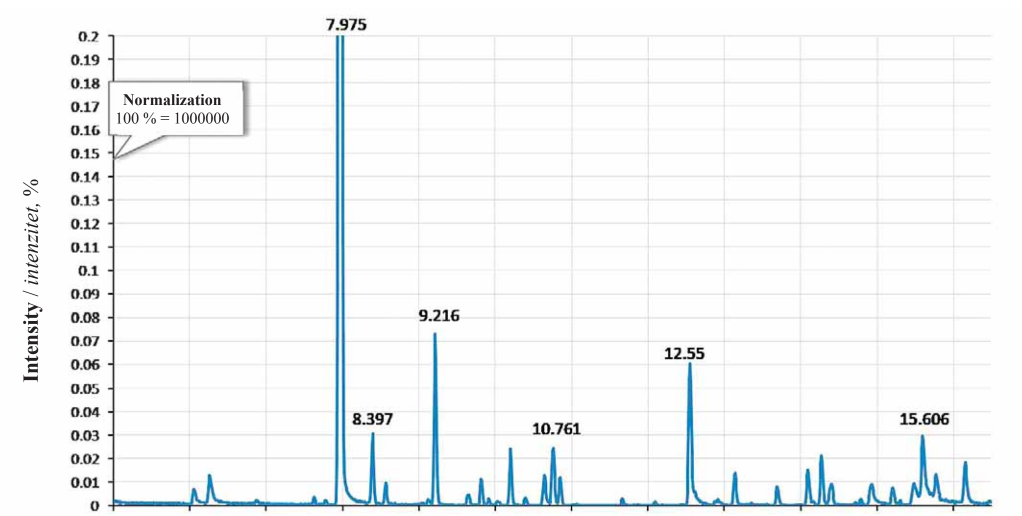

**Figure 1** Chromatogram of PR
**Slika 1.** Kromatogram PR-a

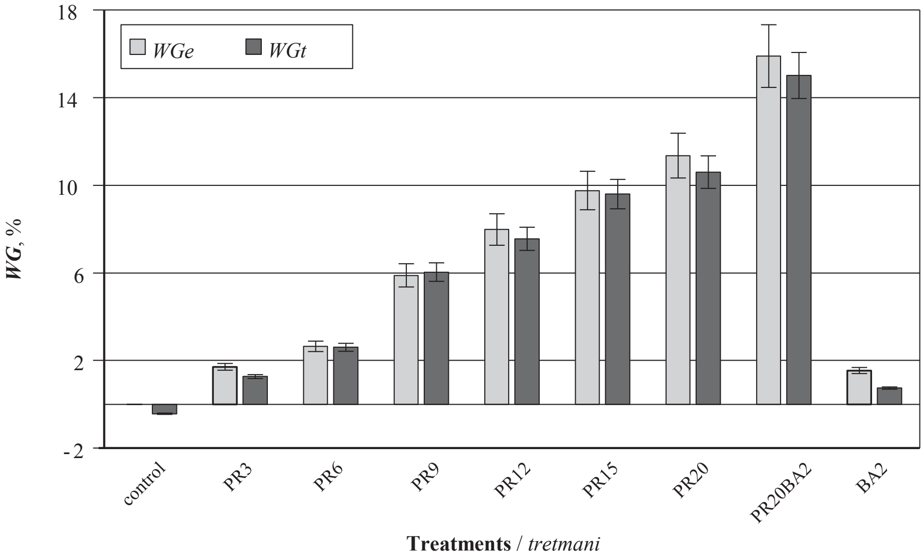

**Figure 2** WG values of treated samples
**Slika 2.** WG vrijednosti tretiranih uzoraka

fungi. However, Beachler and Roth (1956) reported that boric acid retentions between 0.52 and 2.88 kg/m$^3$ are required to protect wood against *G. trabeum* in laboratory conditions. In wood treated with creosote or other oil compounds, higher preservative levels (200 kg/m$^3$) are necessary to prevent biological destruction (Lebow, 2010).

### 3.3 Water absorption and volumetric swelling analyses
### 3.3. Analiza upojnosti vode i volumetrijskog bubrenja

The water absorption of samples from different treatments is shown in Figure 4. At 0-8 h, the water absorption of the control and all treated samples was slow, and the water absorption for all samples was below the FSP. At 8-96 h, the water absorptions increased rapidly until 96-192 h, where they increased slowly. As shown in the figure, the water absorption decreased as the concentration of PR increased. This result is related to the water-insoluble α- pinene in PR and its hydrophobic property (Lebrero, 2012). The water absorption of the 2 % boric acid treated sample is similar to that of the control sample and confirms the leachability of boron in water.

The samples treated with 20 % PR and 20 % PR-2 % boric acid had the lowest water absorption. The water absorption in 20 % PR-2 % boric acid treated sample at 192 h was reduced by 18.69 % in comparison to the control sample.

The volume swelling of the samples is depicted in Figure 5. As can be seen, the volumetric swelling of the control and all treated samples slowly increased at 0-8 h. The volumetric swelling sharply increased at 8-48 h, and then slowly increased after 48 h. This result can be related to the water saturation point of the samples, which does not change very much.

The volumetric swelling of the PR treated samples decreased as the *WG* increased. The maximum volumetric swelling was observed for the untreated controls, and the minimum was related to the PR20 %-BA2 % sample. The decrease in volumetric swelling of the treated samples is probably due to the hydrophobic nature of PR, which may consequently reduce the contact of water with the cell walls (Lebrero, 2012).

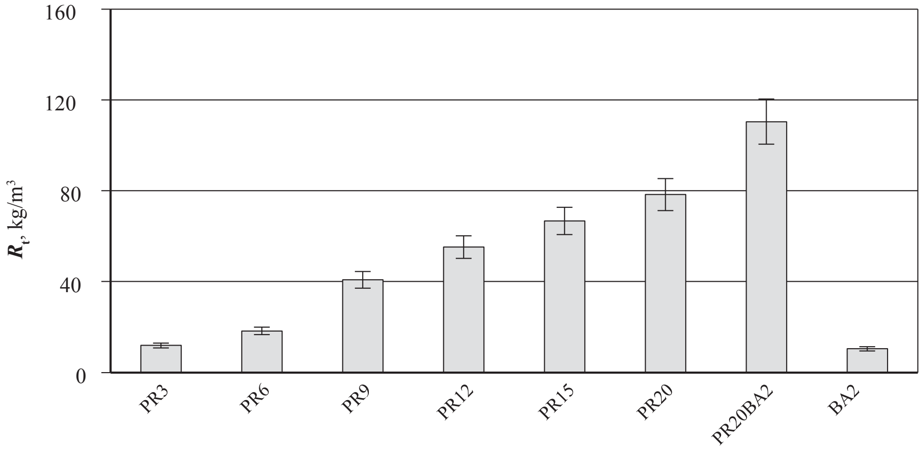

**Figure 3** Theoretical retentions of treated samples
**Slika 3.** Teorijska retencija tretiranih uzoraka

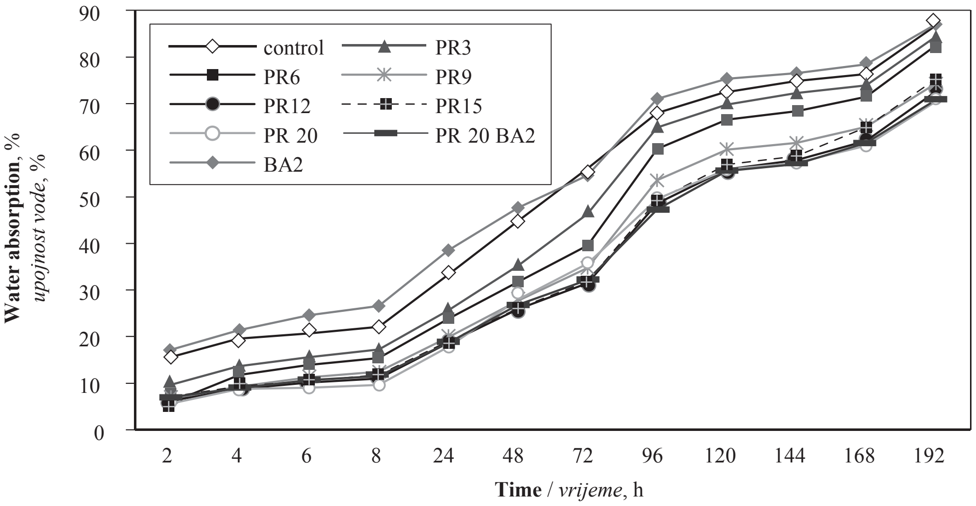

**Figure 4** Water absorptions of samples with time
**Slika 4.** Upojnost vode uzoraka tijekom vremena

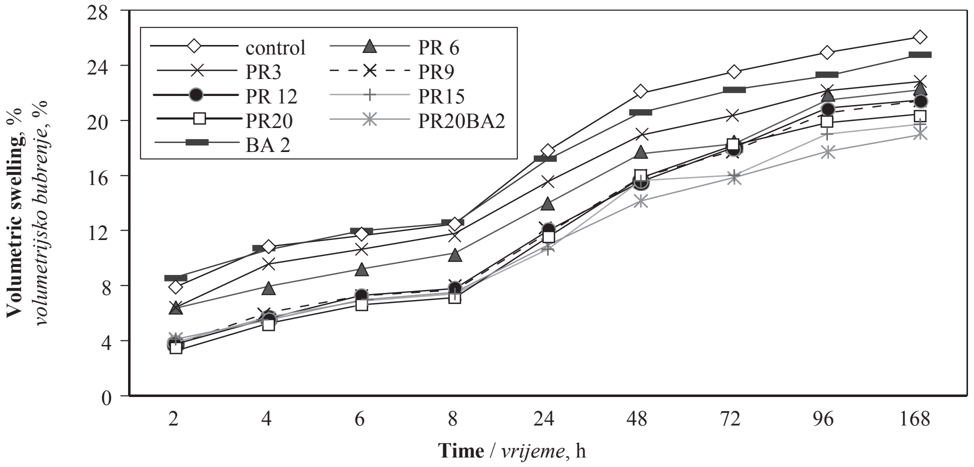

**Figure 5** Volume swelling of samples with time
**Slika 5.** Volumetrijsko bubrenje uzoraka tijekom vremena

It has been reported that the addition of PR into canola oil during the oil heat treatment of poplar wood caused lower volumetric swelling (Mahmoud kia *et al*., 2017).

### 3.4 Leaching analysis
### 3.4. Analiza nakon ispiranja

Figure 6 shows the amount of material released from the treated specimens in a 14 day leaching period. In boric acid treated specimens, the weight loss was 1.86 %, which is almost equal to its weight gain (1.54 %). This result shows that almost all of the boric acid was easily leached from the wood. This agrees with the findings of other researchers (Lebow, 2007; Kartal, 2009). Both PR and dual 20 % PR-2 % boric acid treated specimens showed lesser weight loss as compared to the boric acid treated specimen. About 45 % of the total boric acid remained in the specimens treated with dual PR-boric acid compounds. Lower boric acid leachability in the dual PR-boric acid treated specimen can be related to two reasons: one, less space within the cell wall, which leads to reduction of water absorption, and two, the hydrophobic property of the resin components. The existence of free water in wood provides a suitable environment for fungi growth. Therefore, limiting the amount of free water in wood will reduce fungi attacks. The reduction of water percent gain and volumetric swelling (Figures 4 and 5) emphasizes that the treatment of wood with PR improved its water repellency. Another reason for the decline in leachability of boric acid can be related to the electrostatic interaction of α-pinene and boric acid molecules. α-pinene is an alkene with a carbon-carbon double bond. When it is mixed with boric acid solution, the pi bond between the two carbons is broken. When this pi bond breaks, the sigma bond between the two carbons is still intact, but one of them will have a negative charge and can attract the boron atom in the boric acid molecule. This electrostatic interaction keeps the boric acid in the wood and reduces its leachability in contact with water.

### 3.5 Decay resistance analysis
### 3.5. Analiza otpornosti na propadanje

The results of the fungi test are given in Figure 7. The control samples had a weight loss higher than 20 %, when applying the EN 113 standard criteria. The weight loss of the samples with the 2 % boric acid

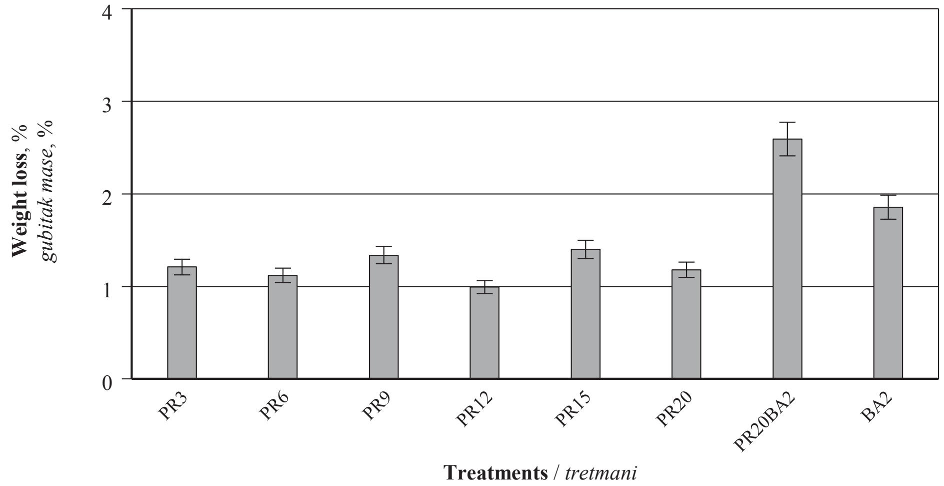

**Figure 6** Weight loss of samples after leaching test
**Slika 6.** Gubitak mase uzoraka nakon ispiranja

treatment was less than 1 % before leaching, but it was high after leaching and almost equal to the weight loss of the untreated samples. The weight loss of the samples saturated with only PR before and after leaching was in the range of 20-35 %, and there was not much difference between the weight loss of the leached and unleached samples. The results showed that, by increasing the PR concentration from 3 to 20 %, the weight loss of the samples against white rot fungi was reduced to less than half compared to the untreated sample, but this is not enough for effective protection of wood according to the EN 113 standard criteria. Previous works also demonstrated the inhibitory effect of the extract of *Pistacia atlantica* on mycelia growth of *Geotrichum candidum* (Talibi *et al.*, 2014).

It was expected that the efficacy of the PR would be related to the chemical composition of the resin. As shown in Table 3, the main components of the resin were α-pinene (85.61 %), α-terpinene (1.94 %), β-pinene (1.72 %), and α-terpinolen (1.32 %). The antifungal property of the resin can be related to the α-pinene, which is the major constituent of PR. α-pinene is a monoterpene made of two structural units of terpenes with five carbon units ($-(C_5H_8)_n$) and a molecular formula of $C_{10}H_{16}$. Studies have shown that terpene compounds, including α-pinene, have analgesic, antibacterial, and antifungal properties. Glisić *et al.* (2007) found that α-pinene is effective against the growth of *Alternaria sp.*, *A. nidulans* and *A. niger*. Salem *et al.* (2016) showed that *P. rigida* oil can inhibit mold growth, which could be related to the major components such as L-α-pinene, α-terpineol, borneol and fenchyl alcohol present in the oil. Powers (2018) outlined that α-pinene-rich (46.1 % α-pinene) commercial *Myrtus communis* essential oil showed a similar antifungal activity against *C. neoformans*.

As shown in Figure 7, the dual treatment of boric acid and PR gave a better effect than boric acid after leaching and better efficiency compared to the PR treatment alone. The weight loss of treated specimens with 20 % PR and 2 % boric acid after leaching was less than 3 %, which provides effective protection against *Trametes versicolor* according to EN 113 standard criteria. This result of the present study showed that boric acid can be successfully fixed in the wood by PR. Moreover, the combination of boric acid and PR improved the fungicide properties of PR.

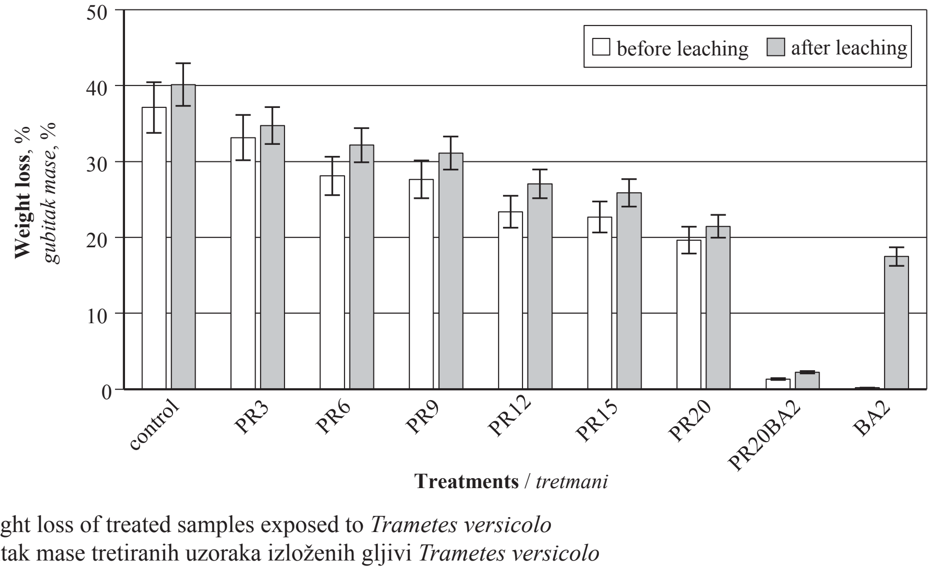

**Figure 7** Weight loss of treated samples exposed to *Trametes versicolo*
**Slika 7.** Gubitak mase tretiranih uzoraka izloženih gljivi *Trametes versicolo*

## 4 CONCLUSIONS
## 4. ZAKLJUČAK

This study was undertaken to investigate the decay resistance performance of beech wood treated with a PR and boric acid mixture against *Trametes versicolor* fungi. PR is an environmentally friendly material that has two fungicide and hydrophobic properties, which can prevent wood decay. Fungi test showed that PR could not fully protect the wood and must be combined with other preservatives. The 20 % PR-2 % boric acid treatment showed a suitable protection against fungi with 2.2 % weight loss. Also, the samples treated with 20 % PR-2 % boric acid showed a 26.9 and 18.69 % reduction in volumetric swelling and water absorption, respectively, in comparison with the untreated sample.

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# Mechanical Properties and Free Formaldehyde Content of Particleboards Produced using Ammonium Sulphate-Based Hardener Partially Replaced with Tartaric Acid

## Mehanička svojstva i sadržaj slobodnog formaldehida ploča iverica izrađenih primjenom katalizatora na bazi amonijeva sulfata djelomično zamijenjenoga vinskom kiselinom

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**ABSTRACT •** *The use of resins, whose curing reaction takes place by high temperature and hardener addition, is inevitably involved in particleboard manufacturing process. In addition to commercial hardeners, such as ammonium sulphate, with the aim of optimizing the production process and reducing the production costs, a certain percentage of hardener can, among other things, be replaced with price affordable bio-based materials. Tartaric acid, that is its salts (tartrates), which are commercially produced for the needs of wine and food industries, are a part of the aforementioned group of products. Since tartaric acid is a relatively inexpensive, readily available, weak diprotic and aldaric acid, the question arises whether it can be used as a component of the hardener system for curing urea-formaldehyde resins that are commercially used in particleboard production. For that reason, in this paper, the influence of partial replacement of ammonium sulphate hardener with tartaric acid on the mechanical properties (bending strength, modulus of elasticity and internal bond) and free formaldehyde content of experimentally produced particleboards was examined. Boards thickness, density and moisture content were also determined. The test results suggest that tartaric acid has a beneficial effect on the above particleboard properties, but they also indicate that the extent of that effect is strongly dependent on panel press time.*

***Key words:*** *particleboards; hardener; tartaric acid; mechanical properties; free formaldehyde content*

**SAŽETAK •** *Proces proizvodnje ploča iverica neminovno podrazumijeva upotrebu smola čija se reakcija otvrdnjavanja ostvaruje uz pomoć povišene temperature i katalizatora. Uz komercijalne katalizatore poput amonijeva*

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*sulfata, radi optimizacije procesa i racionalizacije troškova proizvodnje ploča, katalizatore je u određenom postotku moguće zamijeniti, među ostalim, i cijenom prihvatljivim proizvodima na biobazi. U navedenu se skupinu proizvoda svrstava i vinska kiselina, točnije njezine soli (tartarati), komercijalno proizvedene za potrebe vinske i prehrambene industrije. S obzirom na to da je vinska kiselina relativno jeftina, lako dostupna, slaba diprotonska, aldarna kiselina, postavlja se pitanje potencijala njezine primjene u sustavima katalizatora za otvrdnjavanje urea-formaldehidne smole koja se upotrebljava u proizvodnji ploča iverica. Upravo je zato u ovom radu ispitano kako se zamjena dijela amonijeva sulfata tartaratnom kiselinom odražava na mehanička svojstva (savojnu čvrstoću, modul elastičnosti, vlačnu čvrstoću) i na koncentraciju slobodnog formaldehida eksperimentalno proizvedenih ploča iverica. Usto, u eksperimentu su određene debljina i gustoća ploča te sadržaj vode u njima. Rezultati ispitivanja upućuju na to da dodatak vinske kiseline povoljno utječe na navedena svojstva ploča, ali i na činjenicu kako taj učinak iznimno ovisi o vremenu prešanja ploča.*

***Ključne riječi:*** *ploče iverice; katalizator; vinska kiselina; mehanička svojstva; koncentracija slobodnog formaldehida*

## 1 INTRODUCTION
## 1. UVOD

The urea-formaldehyde (UF) resin is the predominant resin in today's modern particleboard production. This is due to its low price, water solubility, fire resistance, good thermal conductivity, absence of colour of cured resin and its easy adaptation to different curing conditions (Pizzi, 2003). Commonly, UF resins are cured at elevated temperatures using acidic hardeners (catalyst), such as ammonium sulphate ($(NH_4)_2SO_4$). As resin curing depends on the type of hardener, its addition, pH value and solid content (Uner and Olgun, 2017; Xing *et al.*, 2007), the use of various hardeners, other than the above mentioned, is also possible. Such hardeners include ammonium chloride (Xing *et al.*, 2007), sodium chloride, sodium chlorite and borax (Uner and Olgun, 2017), ammonium citrate and zinc nitrate (Park *et al.*, 2003), aluminium chloride and aluminium sulphate (Aizat *et al.*, 2019), ammonium formate for melamine-urea-formaldehyde resins (Čuk *et al.*, 2011) and ammonium persulfate and ferrum (III) chloride if UF resins are used for plywood bonding (Bekhta *et al.*, 2016).

As for the natural acids, attempts were made to use citric and oxalic acids as hardeners in crude state after the wood particles were blended with adhesive mixtures. The use of such hardeners for curing commercial UF resins resulted in a slight rise in the free formaldehyde content (to the upper limit of the E1 emission class). However, if oxalic acid was used neat or in combination with ammonium sulphate for curing low formaldehyde to urea molar ratio UF resin, produced particleboards had high values of internal bond (IB) with free formaldehyde values ≤ 4.5 mg/100 g oven dry board (Costa *et al.*, 2014). The use of citric acid-sucrose mixtures as binders for particleboard production was also attempted. The results showed that although citric acid and sucrose mixtures have great potential as binders for particleboards, the final effect of their use is highly dependent on targeted board density and press temperature. Boards pressed at 200 °C with targeted density of 0.8 g/cm$^3$ had values of mechanical (bending strength (MOR) and modulus of elasticity (MOR)) and physical properties (water absorption and swelling in thickness) in the range or above those minimum required by the applicable standards (Widyorini *et al.*, 2016; Umemura *et al.*, 2013, 2015). As for the low density, when insulating particleboards were pressed using the same binder mixtures, the MOR and swelling in thickness values increased with increasing the board density, and boards with targeted density of 0.4 g/cm$^3$ pressed by adding 15 % binder met the standards requirements (Liao *et al.*, 2016).

Given that our earlier work, in which we have examined the neat resin-catalyst systems (Španić *et al.*, 2017), showed that tartaric acid ($C_4H_6O_6$) could be successfully used as a part of hardener for UF resin curing, and taking into consideration the above mentioned results of other authors, in this paper the experimental particleboards were produced and their mechanical properties and free formaldehyde content were determined. Boards with nominal thickness of 15 mm were made using ammonium sulphate hardener, partially replaced with tartaric acid, with pressing temperature set at 175 °C and pressing time varying from 240 to 300 seconds.

## 2 MATERIALS AND METHODS
## 2. MATERIJALI I METODE

In this study, four series (8 boards in total) of three-layer particleboards sized 45×37×15 (mm) were produced using industrial particles of various wood species and commercial urea-formaldehyde resin. Paraffin emulsion was used as hydrophobic agent and ammonium sulphate-based catalyst partially modified with tartaric acid (1 % addition) was used as hardener. Face and core particles, urea-formaldehyde resin, industrial grade ammonium sulphate and paraffin emulsion were obtained from Kronospan CRO Ltd. Industrial grade tartaric acid (L-(+)-tartaric acid) for oenological purposes was obtained from Begerow GmbH & Co. Deionised water used for catalyst preparation was ASTM type II, and prepared using TKA MicroMed system.

### 2.1 Particleboard production
### 2.1. Izrada ploča iverica

The moisture content for face layer and core layer particles was 7 % and 5 %, respectively, as determined gravimetrically. Such particles were used to make 16 mm thick particleboards with target density of 0.75 g/

**Table 1** Experimental design for produced particleboards
**Tablica 1.** Parametri za izradu eksperimentalnih ploča iverica

| **Panel series**<br>*Serija ploča* | **Press temperature**<br>*Temperatura prešanja*<br>°C | **Press pressure**<br>*Tlak prešanja*<br>N/mm$^2$ | **Pressing time**<br>*Vrijeme prešanja*<br>s | **Hardener type**<br>*Vrsta katalizatora* |
|---|---|---|---|---|
| A | 175 | 3.2 | 300 | neat $(NH_4)_2SO_4$ |
| B | 175 | 3.2 | 300 | $(NH_4)_2SO_4 + C_4H_6O_6$ |
| C | 175 | 3.2 | 270 | $(NH_4)_2SO_4 + C_4H_6O_6$ |
| D | 175 | 3.2 | 240 | $(NH_4)_2SO_4 + C_4H_6O_6$ |

cm$^3$. Panels were made by adding 11 % (face) and 8.5 % (core) of urea-formaldehyde adhesive with 66 % solid content, based on oven dry weight of particles. For the first series of panels (marked A), neat ammonium sulphate was used as hardener, while in other three series (marked B, C and D), the catalyst consisted of ammonium sulphate and tartaric acid. The hardener was added based on the solid content of the resin, 3 % being added for the core layer and 0.2 % for the face layer. In all series of particleboards, solid content of the catalyst was 20 %. In modified catalyst systems, 1 % (based on solid content of resin) of ammonium sulphate was replaced with the same amount of solid tartaric acid, in accordance with the findings of our prior work (Španić *et al*., 2017). Paraffin emulsion solid content was 46 % and was added to face layer (3 %) and core layer (4 %), respectively, based on resin weight.

All series of panels were pressed at 175 °C and 3.2 N/mm$^2$ pressure, with pressing time and hardener used depending on the panel series (Table 1).

In each panel series, two boards were made and conditioned at 65 % relative humidity and 20 °C, before being sawn into test samples.

### 2.2 Particleboard testing
### 2.2. Ispitivanje ploča iverica

Prepared particleboards were tested in order to determine bending strength (*MOR*) and modulus of elasticity (*MOE*) according to EN 310:1993 and internal bond (*IB*) according to EN 319:1993. Mechanical properties were determined using a Schenck Trebel universal testing machine (model UPM 20T). The dimensions of the test samples were determined according to EN 325:1993 using an INSIZE digital calliper (model 1137-150; 0.01mm precision) and an INSIZE digital micrometre (model 3100-25; 0.001mm precision). Board density was determined according to EN 323:1993 and their moisture content using a Memmert laboratory drying oven (model UF 110 plus) according to EN 322:1993. The weight of the test specimens was determined using a Sartorius digital laboratory balance (model TE 612-L; 0.01g precision). Free formaldehyde content was determined according to EN 120:1992, using a Shimadzu UV-VIS spectrophotometer (model UV mini 1240). For each panel series and each property examined, 10 samples were prepared and tested. Free formaldehyde content was determined on one sample per board series.

Data obtained for each property examined was statistically analysed. The influence of partial replacement of ammonium sulphate hardener with tartaric acid on properties of particleboards was evaluated by analysis of variance (ANOVA). Tukey's HSD post hoc test was performed in order to identify which group (data for each property examined for each board series) was significantly different from other groups at 95 % confidence level. Statistical analysis was performed using Statistica Ver.13.3 software (Tibco Software Inc.).

## 3 RESULTS AND DISCUSSION
## 3. REZULTATI I RASPRAVA

The results of mechanical properties of experimentally produced particleboards are given in Table 2, namely their thickness, density and moisture content.

EN 312:2010 defines values of 0.35 N/mm$^2$, 11 N/mm$^2$ and 1600 N/mm$^2$ as minimum requirements for *IB*, *MOR* and *MOE,* respectively, for Type P2 particleboards (boards for interior fitments, including furniture; for use in dry conditions). The same standard permits the value of ≤ 8 mg/100 g oven dry board, for the free formaldehyde content ($CH_2O$). The results in Table 2 show that 1 % replacement of ammonium sulphate with tartaric acid resulted in only minor changes in mechanical properties, if the board pressing time stayed the same (300 s). This is also the case regarding the free formaldehyde, whose content is 6.49 mg/100 g oven dry board for boards from series A and 6.54 mg/100 g oven dry board for boards from series B (Figure 1). Both the boards from series A and B fulfil the minimum requirements, implying that tartaric acid has a positive effect on resin curing if the pressing time is not altered. However, this is not the case regarding the *IB* of boards from series C and D, which were pressed for 270 and 240 seconds, respectively. Although those values are lower, *MOR* and *MOE* values are still above the minimum required. The same is with the free formaldehyde content, which rises up to 7.6 mg/100 g oven dry board for boards from series C and then drops back to 6.62 mg/100 g oven dry board for boards from series D (Figure 1).

Relative increase of MOR and MOE values accompanied by the decrease of IB values of boards hot pressed for 270 and 240 seconds is common. That is due to the fact that the shorter press time influences the relationship between board density and strength, thus leading to higher densification of outer layers, which highly contributes to the increase of MOR and MOE values and decrease of IB values (Moslemi, 1974). More precisely, due to short press time, the heat transfer to inner layers was insufficient, causing improper

**Table 2** Mechanical properties of experimentally produced particleboards
**Tablica 2**. Mehanička svojstva ploča iverica izrađenih za eksperiment

| **Property** *Svojstvo* | **Panel series** *Serija ploča* | *N* | **Mean ± STD** | **Median** | **Min** | **Max** |
|---|---|---|---|---|---|---|
| Thickness / *debljina,* mm | A | 10 | $15.16 \pm 0.15^{ab}$ | 15.15 | 14.97 | 15.43 |
| | B | 10 | $15.22 \pm 0.44^{ab}$ | 15.38 | 14.56 | 15.73 |
| | C | 10 | $15.61 \pm 0.26^{a}$ | 15.42 | 15.31 | 15.96 |
| | D | 10 | $15.15 \pm 0.40^{b}$ | 14.97 | 14.71 | 15.91 |
| Density / *gustoća,* g/cm$^3$ | A | 10 | $0.75 \pm 0.04^{abc}$ | 0.76 | 0.66 | 0.79 |
| | B | 10 | $0.71 \pm 0.03^{c}$ | 0.71 | 0.65 | 0.75 |
| | C | 10 | $0.73 \pm 0.02^{a}$ | 0.73 | 0.70 | 0.78 |
| | D | 10 | $0.77 \pm 0.04^{ab}$ | 0.78 | 0.67 | 0.82 |
| Moisture content / *sadržaj vode,* % | A | 10 | $7.23 \pm 0.26^{a}$ | 7.25 | 6.76 | 7.59 |
| | B | 10 | $7.02 \pm 0.11^{a}$ | 7.02 | 6.87 | 7.22 |
| | C | 10 | $7.21 \pm 0.17^{a}$ | 7.21 | 6.90 | 7.49 |
| | D | 10 | $7.17 \pm 0.15^{a}$ | 7.13 | 6.98 | 7.50 |
| IB / *vlačna čvrstoća okomito na površinu ploče,* N/mm$^2$ | A | 10 | $0.35 \pm 0.04^{a}$ | 0.34 | 0.27 | 0.39 |
| | B | 10 | $0.38 \pm 0.04^{a}$ | 0.40 | 0.31 | 0.45 |
| | C | 10 | $0.16 \pm 0.01^{b}$ | 0.17 | 0.14 | 0.18 |
| | D | 10 | $0.18 \pm 0.06^{b}$ | 0.17 | 0.04 | 0.28 |
| *MOR / savojna čvrstoća,* N/mm$^2$ | A | 10 | $11.9 \pm 1.39^{a}$ | 11.8 | 9.7 | 14.1 |
| | B | 10 | $13.4 \pm 1.38^{ab}$ | 13.2 | 11.6 | 15.9 |
| | C | 10 | $13.9 \pm 1.83^{ab}$ | 14.1 | 11.2 | 16.7 |
| | D | 10 | $15.5 \pm 2.43^{b}$ | 15.3 | 11.7 | 19.0 |
| MOE / *modul elastičnosti savojne čvrstoće,* N/mm$^2$ | A | 10 | $2423.8 \pm 232.02^{a}$ | 2451.3 | 2081.0 | 2699.6 |
| | B | 10 | $2503.7 \pm 256.97^{ab}$ | 2535.6 | 2129.0 | 2892.2 |
| | C | 10 | $2502.7 \pm 256.31^{ab}$ | 2504.2 | 2092.7 | 2857.7 |
| | D | 10 | $2841.0 \pm 345.07^{b}$ | 2881.6 | 2594.2 | 3087.9 |

* Means sharing the same superscript are not significantly different from each other (Tukey's HSD, $p<0.05$)
* *Srednje vrijednosti koje imaju istu slovnu oznaku međusobno se znatno ne razlikuju (Tukey's HSD, p<0,05)*

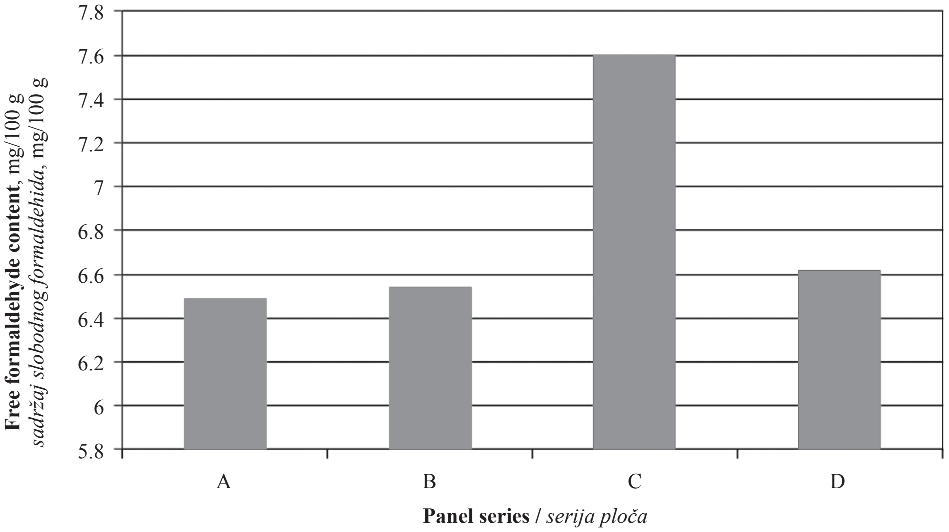

**Figure 1** Free formaldehyde content of experimentally produced particleboards
**Slika 1.** Sadržaj slobodnog formaldehida ploča iverica izrađenih za eksperiment

resin polymerization in the core layers. The slight rise of free formaldehyde content in case of boards from series C and D is also an indication of improper resin polymerization in inner layers.

As already mentioned, the mechanical properties of boards from second series (B) are only slightly affected by the addition of tartaric acid, if compared with the results obtained for the boards made using neat ammonium sulphate (series A). Such results are likely due to the hardener pKa (acid dissociation change) that shifts to lower values as the temperature of wood particle mat increases. With the increase of temperature, tartaric acid pKa changed, thus lowering the pH value of the catalyst system, which resulted in somewhat faster resin curing time. Faster curing consequently helped the development of *IB*, *MOR* and *MOE* values (Wang and Winistorfer, 2003) in case of boards from the second series (B).

The results given in Table 2 reveal that there are no significant differences in moisture content between individual panel series. Small differences in terms of board thickness could be associated with unevenness of metal sheets on which the wet mats were formed. The density drop from 0.75 g/cm$^3$ (series A) to 0.71 g/cm$^3$ (series B) and its rise in series C and D (to 0.73 g/cm$^3$ and 0.77 g/cm$^3$, respectively) could be an indication that, in case of the use of tartaric acid, the catalytic mechanism is independent of the free formaldehyde (Costa *et al*., 2014).

## 4 CONCLUSION
## 4. ZAKLJUČAK

This study showed that it is possible to use ammonium sulphate-based hardener partially replaced with tartaric acid for particleboard production. However, this is possible only if the panel press time is not too short (less than 300 s), as results showed that the IB, MOR and MOE values, as well as free formaldehyde content, are under the influence of set press time. The results also imply that the hardener system and consequently the board properties are influenced by the addition of tartaric acid (pKa values), leading to the conclusion that the particleboards could be produced using neat tartaric acid as hardener for UF resin. Still, this should be examined in further studies.

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# Possibilities of Increasing Renewable Energy in Croatia, Slovenia and Slovakia – Wood Pellets

## Mogućnosti povećanja obnovljivih izvora energije u Hrvatskoj, Sloveniji i Slovačkoj – drvni peleti

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**ABSTRACT •** *Energy from renewable sources is globally a very important issue. In order to reduce pollution and greenhouse gas emissions, many countries enact laws for enhancing the consumption of renewable energy sources. Sooner or later traditional non-renewable energy sources would have to be replaced with alternative energy sources that are already used in developed countries. According to the European Commission (2018), the EU is already a global leader in the sustainable use of natural resources within an efficient bio-economy. The wood pellet industry is becoming more important and the use of wood pellets is increasing as one of the most important contributors to the renewable energy goals of the EU. Wood pellet is an environmentally acceptable product and its production contributes to desirable rural development and sustainable approach to the management of timber resources. This paper focuses on the market of wood pellets used for heating, pinpointing differences in consumers point of view, and drafting the possibilities of future consumption enhancement. Results revealed a relatively low use of wood pellets in all the three surveyed markets. The highest share of consumers of wood pellets came from the areas with less than 5,000 inhabitants. Such households were mostly over 20 years old. The most important factors to decide to use such source of energy were space (required for the heating system) followed by price of woode pellets.*

***Key words:*** *wood industry; wood pellets; renewable energy source; Croatia; Slovenia; Slovakia; households*

**SAŽETAK •** *Energija iz obnovljivih izvora globalno je vrlo važno pitanje. Radi smanjenja onečišćenja i emisij stakleničkih plinova, države promiču zakone o poticanju potrošnje obnovljivih izvora energije. Prije ili kasnije tradicionalne neobnovljive izvore energije bit će potrebno zamijeniti alternativnima, koji se već koriste u razvijenim zemljama. Prema Europskoj komisiji, Europska unija (EU) lider je u održivom iskorištavanju prirodnih resursa*

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*u sklopu učinkovite bioekonomije. U tom kontekstu industrija drvnih peleta, kao i njihova upotreba, iz dana u dan postaju sve nezaobilazniji. U dijelu koji se odnosi na obnovljive izvore energije drvni su peleti jedan od važnijih elemenata koji pridonose ciljevima EU-a. Riječ je o ekološki prihvatljivom proizvodu, a njegova proizvodnja znatno pridonosi ruralnom razvoju i održivom pristupu gospodarenju prirodnim resursima. Ovim se radom želi potaknuti promišljanje kako proizvođača, tako i potrošača drvnih peleta o izrazitom tržišnom potencijalu drvnog peleta za grijanje u hrvatskim, slovenskim i slovačkim kućanstvima. Nadalje, u radu su prikazane razlike u demografskim profilima ispitanika, njihovo znanje o drvenim peletima općenito te o mogućnostima budućeg poticanja potrošnje drvnih peleta u Hrvatskoj, Sloveniji i Slovačkoj. Rezultati su pokazali da je uporaba peleta na sva tri istraživana tržišta relativno niska. Najveći udio potrošnje peleta zabilježen je među potrošačima iz manjih područja (do 5000 stanovnika), a većina stambenih objekata ispitanika stariji su od 20 godina. Najvažniji razlozi zbog kojih se potrošači odlučuju na upotrebu peleta kao izvora grijanja jest prostor potreban za instalaciju opreme za grijanje na pelete te cijena peleta.*

***Ključne riječi:*** *drvna industrija; drvni pelet; obnovljivi izvori energije; Hrvatska; Slovenija; Slovačka; kućanstva*

## 1 INTRODUCTION
## 1. UVOD

Energy from renewable sources is globally a very important issue. In order to reduce pollution and greenhouse gas emissions, many countries enact laws for enhancing the consumption of renewable energy sources. For many years, the European Union has been one of the world leaders in the promotion of renewable energy trying to change the relations in energy, favoring renewable energy through applicable laws and active implementation of incentive programs, simultaneously providing generous subsidies. More precisely, the aim of the European Union (EU) as a whole is to provide 20 % of energy from renewable energy sources for its gross energy consumption by 2020 and 32.5 % by 2030 (Annual activity reports, 2019). Sooner or later, traditional non-renewable energy sources would have to be replaced with alternative energy sources that are already used in developed countries. According to the European Commission Report (2018), the EU is already a global leader in the sustainable use of natural resources within an efficient bio-economy. The bioeconomy covers all sectors and systems that rely on biological resources, their functions and principles. It includes and interlinks: land and marine ecosystems and the services they provide; all primary production sectors that use and produce biological resources (agriculture, forestry, fisheries and aquaculture); and all economic and industrial sectors that use biological resources and processes to produce food, feed, bio-based products, energy and services. In other words, it implies a shift from fossil resources (oil) to renewable resources (biomass) with an accent on the development and production of new products from biomass that must be implemented in a sustainable manner (Šupín and Dzian, 2018). A lot of studies from central and southern Europe dealing with forest-based sector emphasize the role of wood and wood products within the European Green Deal (Dudík *et al*., 2019; Glavonjić *et al*., 2015; Kaputa *et al*., 2017; Maťová and Kaputa, 2018; Olšiaková *et al*., 2016; Paluš *et al*., 2014; Paluš *et al*., 2018; Parobek *et al*., 2016; Pirc Barčić *et al*., 2015; Pirc Barčić *et al*., 2019; Potkány and Debnár, 2018; Šupín, 2015).

The majority of demand for pellets originates from the European Union (EU) (precisely 30.3 % of the total world production, according to the Calderón and Colla, 2019) in response to its greenhouse gas (GHG) (NREL, 2013) and emission mitigation policy, as a result of the EU objective to increase the share of renewable energy. The wood pellet industry and the use of wood pellets is now becoming more and more important. Moreover, wood pellets are emerging as one of the most important contributors to the renewable energy goals of the EU. Flinkman *et al.* (2018) stated that wood pellets represent the best international and dynamic character of the European renewable energy market. Furhtermore, when talking about ecological and ecotoxicological aspects, the potential role of wood-pellets in a sustainable biofuel system is an important issue (Olsson *et al.,* 2011).

### 1.1 Wood pellets as eco-acceptable source
### 1.1. Drvni peleti kao ekološki prihvatljiv izvor energije

Wood-pellet is a renewable fuel with no carbon dioxide emissions that would contribute to global warming, and with the highest potential to reduce carbon dioxide (Wahlund *et al.*, 2004). Additionally, as defined in ENplus standard (2015), wood pellets are a woody biofuel shaped in a cylindrical form with random length, typically 3.15 to 40 mm, with a diameter of about 6 or 8 mm, and broken ends. The advantage of wood pellets, compared to the original biomass, lies in a higher energy density, homogeneous quality, improved handling and storage properties, and better applicability for different end uses. According to Sikkema *et al*. (2011) and Flinkman *et al*. (2018), two main categories and related markets can be identified: i) industrial grade pellets for large scale power production and ii) residential grade (or non-industrial pellets for small-scale to medium-scale production) pellets. The production of wood pellets started in the 1970s in the United States and Canada, while the first large scale production took off in the 1990s, also in the US and Canada (Peksa-Blanchar *et al.,* 2007; Sikkema *et al.,* 2011; Thrän *et al.,* 2018). In Europe, the first developments were recorded in Sweden in the 1980s as a result of the second oil price/energy crisis. However, in Sweden in 1992, the wood pellets market started to be competitive almost overnight, after the introduction of fiscal measures in taxing fossil fuels. The wood pellets markets started to develop in Austria, Denmark, Italy, and Germany in the late 1990s (Peksa-Blanchar *et al.,* 2007). Since that time, wood pellets markets have been expanded and transformed (Thrän *et al.,* 2018). In 2011, Cocchi *et al.* presented the first compilation of the global production and trading of wood pellets for energy with data from 31 countries in the Americas, Europe, and Asia, showing an estimated increase in consumption of about 110 % between 2006 and 2010, reaching 13.5 million tones. The major import originates from North America but the increasing demands have stimulated advances in Russia, Africa, South America and Asia (Verhoest and Ryckmans, 2012). Since that time, markets have been further expanded and diversified. Experts from the International Energy Agency (IEA) Bioenergy Task 40 have collected and analyzed the market situation in all wood pellet related countries. IEA Bioenergy Task 40 focuses on sustainable biomass markets and international trade to support the biobased economy. As both production and consumption increase, trade in pellets is becoming more significant and the market for this new fuel is becoming increasingly global, with imports to Europe from as far as western Canada and China.

Besides the development of the countries' policies, demands, and supply patterns (Thrän *et al.,* 2018), papers mostly deal with trade flow trends, apparent consumption (including industrial and non-industrial wood pellets consumers), and pellets producers in the context of facility capacities. There is, however, a lack of information about the profiles of non-industrial wood pellet consumers. Flinkman *et al.* (2018) noted that a questionnaire sent to wood pellet companies indicates that specific state subsidies could be a driver for the purchase of pellet stoves and boilers, resulting in a basic level of consumption of non-industrial pellets. Further, light heating oil and natural gas are considered the main heating sources replaced by wood pellets.

The question is how to describe the profiles of wood pellet consumers and types of their households. Given that the development of wood pellets market has been very country-specific, also driven by national fiscal and financial incentives to influence their own domestic market (Dicken, 2008), the scope of this paper was to explore how much Croatian, Slovenian and Slovak households know about wood pellets in general and what the possibilities of their future consumption enhancement are. By signing the Kyoto Protocol, Croatia, Slovenia and Slovakia have committed themselves to reduce greenhouse gas emissions by five percent by 2020 and to promote the use of wood biomass, especially wood pellets, this being one of the activities required to meet these obligations. This paper sheds light on the situation and answers the questions about the wood pellets market potential for heating, pinpointing differences in the demographic profiles of the respondents, their knowledge about wood pellets in general and the possibilities of their future consumption enhancement.

## 2 MATERIALS AND METHODS
## 2. MATERIJALI I METODE

For this research, the following non-industrial pellets markets were selected: Slovenia, Croatia and Slovakia. These counties have a high potential of renewable energy sources. In order to collect consumer data for this research, a telephone survey was used, with a total of 1916 respondents. The first survey was conducted in Croatia in 2015 (preliminary research done by Pirc Barčić *et al.*, 2015), the second in 2017 (Slovenia and Croatia market), and the third one in 2018 for the Slovak market. Regarding the samples by countries, 915 respondents were from Slovenia, 887 from Croatia, and 225 from Slovakia. In all the three markets, identical questions were used. This non-specific population was targeted because there were no records on consumer consumption of renewable sources, which could narrow down the target population.

A telephone survey was used for surveying respondents for this study. This approach was selected because, according to Roster *et al.* (2004), it is possible to obtain a representative national sample by telephone surveys. The relative advantages of telephone surveys, like lower cost, less risk of interviewer bias, and avoidance of cluster sampling, were additional elements in making decision about the method of data collection (Berrens *et al.*, 2003).

Based on research objectives, a questionnaire was developed, pre-tested, and finalized based on pretested inputs. Straightforward questions and Yes/No items were used. Furthermore, multi choice item measure was used because, according to Thorndike (1967) cited by Lewis-Beck *et al.* (2004), it can be superior to a single, straightforward question. At the beginning of the phone interview, the researcher gives an introductory statement presenting the research study, introduces himself and explains the respondent's role emphasizing that his/her participation in research is essential (Dillman *et al.*, 1976) was presented to respondent.

The questionnaire consisted of twenty-one open questions, asking respondents about their houshold mountly income, types of their households, heating sources in their households, reasons for using wood pellets, availability of pellets price and consumption, other possible factors that could infulence the pellet consumtion, etc. Data were analyzed using descriptive statistics, $\chi2$- test, Fisher's Exact Test, Shapiro-Wilk normality tests, with the use of the SPSS statistical packages.

## 3 RESULTS AND DISCUSSION
## 3. REZULTATI I RASPRAVA

### 3.1 Demographic profile of respondents
### 3.1. Profil ispitanika

Among the considered market population, in all the three countries, households with two (2) and one (1) employees (50.0 % and 22.0 %, respectively) dominate. Observing the population in the area where the households are located, survey results showed that

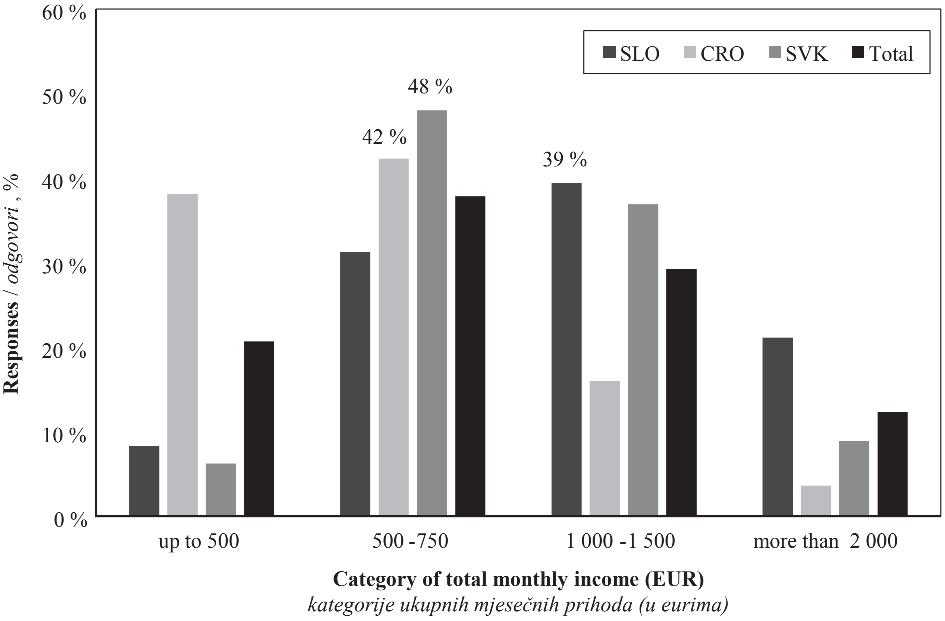

**Figure 1** Total monthly household income per person
**Slika 1.** Ukupni mjesečni prihodi kućanstva po osobi

most households are located in the area with less than 5,000 inhabitants (48.5 %) and from 500,000 to 1,000,000 inhabitants (20.0 %). Regarding their monthly household income per person (Figure 1.), for instance, in Slovenia, the highest precentage (39 %) of the oberved population have an income of 1,000 to 1,500 EUR. In case of Croatia and Slovakia, most respondents stated to have from 500 to 700 EUR (42.0 % and 48.0 %, respectively). Regarding the education level, most respondents declared to have high school graduate and college or university level (61.2 % and 24.5 %, respectively).

Such description needs a few comments. Considering the location of respondents, all the three countries belong to the group of central and south EU countries with rather smaller population (from 2 up to 5.5 million of inhabitants) and no bigger cities (except their capitals). At this point of view, they are comparable. The differences in monthly household income are much more robust – especially in the case of Slovenia with prevalently higher income categories. Comparative reflections on Slovakian and Croatian economies and wood sector can be found in the study of Kaputa *et al*. (2018).

### 3.2 Household characteristics
### 3.2. Obilježja kućanstava

In this research, households more than 20 years old (61.3 %) mostly dominated, followed by those between 10 and 20 years old (24.7 %). It is interesting to note that just 4.5 %, (mainly in Slovenia – 7.1 %) are newer houses, meaning that they are less than 5-year-old.

The characteristics discerning the markets from one another were investigated applying Chi-Square statistic. Tests showed that there is a statistically significant difference between the countries mean values of almost all heating sources, ($\chi^2$ (12) = 720,505, $p$<0.001). As shown in Table 1, on average, the use of firewood as a heating source is the highest (46.4 %, especially in Croatia and Slovenia). Furthermore, there is a light difference when observing the countries individually; for example the Slovak household, where a

**Table 1** Heating sources and heating area of households observed ($n$ = 1966)
**Tablica 1.** Izvori grijanja i grijana površina promatranih kućanstava ($n$ = 1966)

| **Energy source used for heating, %** / *Izvor energije za grijanje,* % | | | | | | | |
|---|---|---|---|---|---|---|---|
| Market<br>*Tržište* | Gas<br>*Plin* | Oil<br>*Ulje* | Electricity<br>*Struja* | Fireod<br>*Ogrjevno drvo* | Solar<br>*Solarni izvori* | Heathing plant<br>*Toplinske pumpe* | Other<br>*Drugo* |
| SLO | 12.1 | 15.3 | 13.8 | 43.3 | 4.1 | 3.1 | 8.3 |
| CRO | 25.2 | 2.5 | 13.5 | 55.1 | 0.3 | 2.5 | 0.9 |
| SVK | 23.6 | 0.0 | 6.2 | 28.9 | 0.4 | 38.2 | 2,7 |
| Total / *Ukupno* | 18.2 | 8.9 | 13.0 | 46.4 | 6.0 | 2.5 | 4.9 |

| **Heating area in households**, % / *Grijana površina,* % | | | | |
|---|---|---|---|---|
| | Up to 50 m$^2$ | 50-100 m$^2$ | 100-200 m$^2$ | >200 m$^2$ |
| SLO | 7.8 | 32.6 | 46.0 | 13.6 |
| CRO | 28.6 | 51.5 | 18.1 | 1.8 |
| SVK | 6.7 | 61.3 | 24.9 | 7.1 |
| Total / *Ukupno* | 16.4 | 43.9 | 31.8 | 7.9 |

*SLO – Slovenia / *Slovenija*; CRO – Croatia / *Hrvatska*; SVK – Slovakia / *Slovačka* heating plant is most frequently the energy source, accounting for 38.2 %. At the other end of the spectrum lie other sources (like heat pump, biomass, briquettes, etc.) and heating plant, only accounting for 3.1 % (Slovenia) and 2.5 % (Croatia). Chi-Square test also showed statistically significant differences in all the three above mentioned cases ($\chi^2$ (6) = 371,460, $p<0.001$). Consequently, in most cases, the results show that the heating area covers 50 to 100 and 100 to 200 square meters (Table 1). Regarding the question about annual heating costs, for testing the differences in the markets, besides $\chi^2$ test, a post-hoc test was made by pairwise comparisons of countries, as well as the applied Bonferroni correction for multiple tests. The tests showed that there is a statistically significant difference between Slovenia and Croatia ($p<0.001$, Fisher exact test) and between Slovenia and Slovakia in annual heating costs ($p<0.001$, Fisher exact test). However, the difference between Croatia and Slovakia were not significant ($p=0.022$, Fisher exact test). Expenses in all the three countries were mostly up to 700 EUR (48.2 %) and ranged between 700 and 1,300 EUR per year (41.1 %). However, when comparing the expenses of heating with the energy source used for heating on annual basis, individually per market, then Slovenian householders pay the highest price, more than 1,300 EUR, for plant heating. On the Croatian and Slovak market, this was the case for oil heating.

### 3.3 Pellet users in Slovenia, Croatia and Slovakia
### 3.3. Korisnici peleta u Sloveniji, Hrvatskoj i Slovačkoj

As already mentioned, wood pellets have an important role for the renewable EU energy goals, in the transformation of the energy system from fossil fuels, which is required to meet the EU 2030 and COP21 targets. To this end, a set of open questions was used to explore how much Slovenian, Croatian, and Slovak households know about wood pellets in general, and what the possibilities of their future consumption enhancement are. The Fisher exact test proved that there is a statistically significant difference between the Slovenian and Croatian population in their familiarity with the term wood pellet ($p<0.001$), more precisely 83.2 % of respondents from Slovenia and 65.3 % from Croatia declared to know about it. The same answer came to the question on their familiarity about lower price of pellets versus other heating sources, such as oil.

Several tests were done to investigate the use of pellets as a main source of heating in households of the observed markets – Slovenia, Croatia and Slovakia. Chi-square test has shown that there is a significant difference between pellets users and non-pellets users, generally ($\chi^2$ (4) = 18,435, $p<0.05$, Pearson Chi-Square). Considering the whole sample ($n$=1,973 respondents) only 6.0 % of respondents stated to use wood pellets as a heating source. Dealing with individual countries, the highest share of (pellets) users were in Slovenia (11.2 %). The data shows a relatively low use of pellets in all the three markets. Among the considered population, the highest percentage of pellets users came from an area with less than 5,000 inhabitants (52.9 %) and with households more than 20 years old (45.4 %). Heating surface mostly (43.7 %) covers from 50 to 100 and from 100 to 200 square meters. Furthermore, such households mainly have 4 persons, where usually only 2 persons work (63.0 %). Monthly household income per person differs among countries. In Slovenia, pellets users receive over 2,001 EUR (38.8 %), in Croatia from 500 to 1,000 EUR, and in Slovakia from 1,001 to 2,000 EUR (30.8 %). These findings point at different respondents' living standards. The majority of respondents (>80 % of answers) are familiar with wood pellets producers in their country, as well as with the price of wood-pellets and government subventions.

Furthermore, the following set of variables was applied to explore the reasons for using pellets: *price, government subventions, ecological aspects, space, decrease of energy, higher heat emission, and long-term cost effectiveness* (Table 2). Fisher's Exact test has shown that there is a statistical difference between respondents across the three markets and regarding the variable *price* (p=0.005). To see the difference between groups, further post hoc tests were done. A Bonferroni correction has been applied for Chi square test on comparison of groups. With four pairwise comparisons, p-value must be less than 0.05/6=0.008 to be significant at $p<0.05$ level. The results indicated a higher share of Slovak respondents, who marked price as the reason for using wood-pellets (Cramer's V=0.322, $p<0.001$). Even though there were no statistically significant differences between other variables, when observing the results of descriptive analysis (Table 2), it can be seen that the most important reasons for the use of pellets are *space* and then *price* (66.3 % and 33.7 %, respectively). At the end of the spectrum was a government subvention (4.2 %). There are no differences among the individual markets. For instance, in case of Slovenian users, the most important reason was *space* (67.5 %), while in case of Slovakian users, it was *price* (77.8 %).

To explore the benefits of pellets, the participants were asked to estimate annual heating costs and their satisfaction with the amount of produced heat. The Fisher's Exact Test was applied to investige the characteristics discerning the three markets from one another. The results showed that there are3 statistically significant differences between respondents and annual wood-pellet costs ($p=0.040$) across all the three markets. The annual heating costs for pellets mostly ranged around 1,000 EUR in case of Slovenian users (75.6 %) and Slovak users (66.7 %). The majority of Croatian users (66.7 %) pay the price between 1,000 and 2,000 EUR. Comparing this result with non-pellets users, the costs are slightly lower for pellets users. Their satisfaction with the amount of produced heat is not statistically significant ($p>0.05$), but generally almost all the pellets users (94.6 %) expressed satisfaction with this heating option.

The research also focused on the source of information about pellets. Respondents indicated **media**

**Table 2** Reasons for using wood-pellets and annual wood-pellets costs (in EUR) on observed markets ($n$ = 119)
**Tablica 2.** Razlozi odabira peleta i godišnja cijena takvoga grijanja na promatranom tržištu ($n$ = 119)

| | **Reasons for using wood-pellets, %** / *Razlozi odabira peleta,* % | | | | | | |
|---|---|---|---|---|---|---|---|
| Market *Tržište* | Price *Cijena* | Government subventions *Državne subvencije* | Ecological aspects *Ekološki aspekti* | Space *Prostor* | Decrease of energy use *Smanjenje potrošnje energije* | Higher heat emisssion *Veća toplinska energija* | Long-term cost effectiveness *Dugoročna isplativost* |
| SLO | 30.1 | 4.8 | 27.7 | 67.5 | 26.5 | 22.9 | 7.2 |
| CRO | 0.0 | 0.0 | 0.0 | 100.0 | 0.0 | 0.0 | 0.0 |
| SVK | 77.8 | 0.0 | 11.1 | 44.4 | 0.0 | 11.1 | 0.0 |
| Total YES *Ukupno DA* | 33.7 | 4.2 | 25.3 | 66.3 | 23.2 | 21.1 | 6.3 |
| | **Annual pellets costs (in EUR), %** / *Godišnji troškovi peleta (u eurima),* % | | | | | | |
| | Up to 1 000 /*do 1 000* | | | 1 000-2 000 | | >2 000 | |
| SLO | 62.2 | | | 19.4 | | 0.0 | |
| CRO | 0.0 | | | 33.3 | | 0.00 | |
| SVK | 38.5 | | | 23.1 | | 7.7 | |
| Total / *Ukupno* | 57.1 | | | 21.0 | | 0.8 | |

*SLO – Slovenia / *Slovenija*; CRO – Croatia / *Hrvatska*; SVK – Slovakia / *Slovačka*

(33.7 %) as the main source of information about wood pellets, followed by some other communication channels (25.3 % of responses). Recommendation was of low importance. To determine the difference between markets, a set of Fisher's exact tests were run. These tests showed that there is a statistically significant difference only among respondents across all the three markets and media ($p$=0.005, Cramer's V=0.322, $p$<0.001).

### 3.4. Potential pellets users in Slovenia, Croatia and Slovakia
### 3.4. Potencijalni korisnici peleta u Sloveniji, Hrvatskoj i Slovačkoj

To explore the possibilities of enhancement of wood pellets future consumption, a list of 7 variables was introduced to respondents for evaluation. Table 3 summarizes respondents' statements, validated by chi-square and post hoc tests. The tests showed a statistically significant difference among countries and their intention to use wood-pellets in the future - *within 1 year* ($\chi^2$ (2) = 10.887, $p$<0.05, Pearson Chi-square test). Also, statistically significant difference was revealed among countries and their intention to use wood-pellets in the future - *within 3 years* ($\chi^2$ (2) = 17.744, $p$<0.05, Pearson Chi-square test). The Bonferroni correction has been applied for chi-square test in comparison of groups for these question. With four pairwise comparisons, p-value must be less than 0.008 (0.05/6=0.008) to be significant at the $p$<0.05 level. The results showed a statistically significant difference indicating a higher proportion of Slovenian and Croatian respondents (>94.0 %) who missed to express intention to use pellets within three years (Cramer's V=0.098, $p$<0.001.). As shown in Table 3, most of respondents (11.1 %) stated the possibility to move to start using pellets heating within five years. On other side, no possibility to install wood pellets heating system in household was seen as the most important barrier (27.8 %) for non-pellets users. This finding is contrary to the response of pellets users since they consider price as a positive characteristic.

**Table 3** Intention to use pellets in future ($n$ = 1598)
**Tablica 3.** Namjera o budućoj uporabi peleta ($n$ = 1598)

| | **Intention to use pellets in future – YES, %** *Namjera o budućoj uporabi peleta – DA,* % | | | **Intention to use pellets in future – NO**, % *Namjera o budućoj uporabi peleta – NE,* % | | | |
|---|---|---|---|---|---|---|---|
| Market *Tržište* | YES – within 1 year *DA – za 1 god.* | YES – within 3 years *DA – za 3 god.* | YES – within 5 years *DA – za 5 god.* | NO – high price *NE – visoka cijena* | NO – insufficient information *NE – nedovoljno informacija* | NO – there is no possibility to install wood pellets heating system in my haousehold *NE – u mojem kućanstvu ne postoji mogućnost instalacije grijanja na pelete* | NE – other *NE – drugi razlozi* |
| SLO | 4.2 | 5.2 | 12.8 | 16.1 | 17.5 | 21.2 | 22.4 |
| CRO | 2.2 | 1.9 | 9.5 | 10.4 | 30.1 | 25.7 | 21.4 |
| SVK | 0.5 | 0.9 | 12.0 | 0.0 | 23.5 | 56.3 | 6.6 |
| Total *Ukupno* | 2.9 | 3.2 | 11.1 | 11.3 | 24.2 | 27.8 | 19.9 |

*SLO – Slovenia / *Slovenija*; CRO – Croatia / *Hrvatska*; SVK – Slovakia / *Slovačka*

Chi-square test also proved statistically significant differences among the observed markets for non-user subgroup that gave a negative answer about the future use of pellets, as presented in Table 3. Statistically significant differences showed that the reasons were *price* ($\chi^2$ (2) = 36.787, $p<0.05$, Pearson Chi-square test), *insufficient information* ($\chi^2$ (2) = 26.868, $p<0.05$, Pearson Chi-square test) and *inadequate technical prerequisites* ($\chi^2$ (2) = 88.183, $p<0.05$, Pearson Chi-square test).

## 4 CONCLUSION
## 4 ZAKLJUČAK

Wood pellets require to be observed from the economic, environmental and social point of view due to the pronounced advantages and volume of raw material resources in Croatia, Slovenia and Slovakia. In many cases, they represent the fuel of the future.

In Croatia, Slovenia and Slovakia, the observed households (48.5 %) are mainly located in the area with less than 5,000 inhabitant, usually generating monthly household income from 500 to 750 EUR. Around 61 percent of households are more than 20 years old and the majority of them use wood as the heating source (46.4 %, especially in case of Croatia and Slovenia). Further, there were some differences when the countries were observed separately. In Slovakia, the most frequent source of heat comes from heating plants (38.2 %), while up to 29.0 % of households use firewood for this purpose. This fact has a historical explanation since most of urban blocks, built during the socialist period, were heated centrally. On the other side, households in rural areas used firewood or gas, or a mix of these two. Such Slovakian households have the biggest potential to become the users of wood pellets.

Heating costs in all the three countries achieved either up to 700 EUR (48.2 %) or between 700 and 1 300 EUR per year (41.1 %). Slovenian householders pay the highest price, more than 1,300 EUR. A statistically significant difference was found between Slovenian and Croatian population in their familiarity with the term wood pellets ($p<0.001$) (83.2 % respondents from Slovenia and 65.3 % from Croatia declared to know about it), but the results revealed a relatively low use of pellets in all the three markets. Additionally, among the considered population, the highest percentage of pellets users came from areas with less than 5,000 inhabitants and with households more than 20 years old. The majority of respondents in all the three countries stated that they are familiar with subventions and government programs that encourge the use of renewable energy sources, but the most important reasons for the use of wood pellets were *space* and then *price,* while at the end of the spectrum was a government subvention (only 4.2 %). This study also showed statistically significant differences between countries and their positive intention to use wood-pellets in the future - *within 1 years* and *within 3 years*. The most frequent reason not to plan to use wood pellets was *no possibility to install wood pellets heating system* in household (27.8 %).

Croatian consumers (public institutions and private persons), similarly as Slovenian and Slovakian consumers, are not informed enough about the scope of use of wood pellets and their advances within the Green Deal. In many cases, the market price, which seems to be perceived as high by consumers, plays the main role. Over all, European Union bioenergy policies, oil prices and sales of heating appliances can have an important effect on wood pellets consumption market. Additionally, segments related to energy efficiency in flats, houses and other buildings can also influence the wood pellets demand, as well as the development of wood-pellets production technologies. Finally, the authors hope that this study will support decision making of wood pellets producers, wood-pellets consumers as well as policy makers to strengthen the development of this wood-based sector in all the surveyed countries.

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# Weathering Performance of Beech Wood Coated with Acrylic Paint Containing UV Stabilizers of Dihydroxy Benzophenone and Nano Zinc Oxide

## Svojstva bukovine obrađene akrilnim premazom koji sadržava UV stabilizatore dihidroksi-benzofenon i nano cinkov oksid nakon izlaganja vremenskim utjecajima

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**ABSTRACT** • *In this study, the effect of UV stabilizers (dihydroxy benzophenone and nano zinc oxide) on the weathering degradation of water-based acrylic coating on beech wood was investigated. The wood specimens were coated by brush and then weathered naturally for six months. The obtained results showed that the use of nano zinc oxide reduced color changes and mold growth on the surface of weathered samples. However, the results of contact angle, pull-off adhesion, colorimeter and FTIR revealed that the dihydroxyl benzophenone was not effective in preventing weathering degradation of coated wood.*

***Keywords:*** *acrylic coating; weathering; beech wood; dihydroxy benzophenone and nano zinc oxide*

**SAŽETAK** • *U radu se prikazuje istraživanje utjecaja UV stabilizatora (dihidroksi-benzofenona i nano cinkova oksida) na razgradnju vodenoga akrilnog premaza na bukovini koja je bila izložena vremenskim utjecajima. Uzorci drva premazani su kistom, a potom su šest mjeseci prirodno izloženi vremenskim utjecajima. Dobiveni su rezultati pokazali da nano cinkov oksid smanjuje promjenu boje i pojavu plijesni na površini izloženih uzoraka. Međutim, rezultati kontaktnog kuta, adhezije, kolorimetrije i FTIR analize potvrđuju da dihidroksi-benzofenon nije učinkovit u sprečavanju razgradnje premazanog drva.*

***Ključne riječi:*** *akrilni premaz; izlaganje vremenskim utjecajima; bukovina; dihidroksi-benzofenon; nano cinkov oksid*

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## 1 INTRODUCTION
## 1. UVOD

Wood possesses several attractive properties, such as aesthetic appeal, low density and mechanical strength, making it one of the most widely used materials for engineering and structural application. Unfortunately, when wood is exposed outdoor, a variety of weathering factors degrade the main wood components (Macleod *et al*., 1995). The factors contributing most to wood degradation by weathering are UV radiation and water; thereby, mainly lignin is decomposed to lower-molecular-weight compounds, which are washed out by rain. Since the penetration of UV waves is superficial, weathering affects the aesthetic properties of wood more than the mechanical ones (Feist, 1982). The change of wood color is the first sign of photo degradation when wood is exposed to UV radiation. Undesirable changes, such as discoloration, roughness and cracks, are observed when wood is further exposed to environmental conditions, such as heat, water and microorganisms (Turkoglu *et al*., 2015, Temiz *et al*., 2007). Different approaches are pursued to protect wood from weathering degradation, whereas the common method is the application of coatings (Bulian and Graystone, 2009; Chang and Chou, 2000). Clear coating is one of the desirable and effective methods, which protects the wood surface keeping the wood grain and color visible, with no negative effect on the wood natural appearance. Water based acrylic polyurethane - as one of the common clear coatings - is durable, highly efficient, non-toxic, and widely applied in the recent years due to growing environmental concerns (Saha *et al*., 2011). For outdoor applications, incorporation of organic and inorganic UV absorbers and hindered amine light stabilizers (HALS) is necessary due to the development of transparent coatings. These compounds are used for the retardation and elimination of photochemical processes in polymers (Malanowski, 2009). The mechanism of stabilizing organic UV absorbers involves absorption of UV light and a subsequent quick dissipation of absorbed energy in the form of harmless long-wavelength radiation. Derivatives of hydroxyl phenyl benzotriazole, hydroxyl phenyl benzophenone and hydroxyl phenyl-s-triazines are applied as organic UV absorbers. Inorganic UV absorbers protect a polymer by reflecting the harmful UV light. Among the inorganic UV absorbers, zinc oxide has a long history of color protection. It has the advantage of a higher photo permanence compared to organic stabilizers (Saha *et al*., 2011; Wang and Tooley, 2011). Furthermore, its effectiveness in blocking UV radiation boosted in nanosized when compared to bulk. However, nanoparticles tend to agglomerate due to their large surface area and high surface energy. The formation of agglomerates reduces the UV absorbing capacity and, therefore, the UV shielding properties of these nanoparticles (Becheri *et al*., 2008). They have to be re-dispersed, most effectively by ultra-sonication, and stabilized to prevent a new agglomeration (Fufa *et al*., 2012). Some studies have shown that the use of zinc oxide (ZnO) as a UV absorber in coatings improves weathering performance of coated wood samples. Weichelt *et al*. (2011) studied the efficiency of ZnO-based acrylate coatings on wood during artificial weathering. Their results show that nano-ZnO acts as a UV absorber by reducing yellowing and improving optical properties. Salla *et al*. (2012) mentioned that the incorporation of ZnO nanoparticles in PU coatings enhanced the photostability of coated rubber wood during weathering.

This study aimed to compare the efficiency of dihydroxy benzophenone and ZnO nanoparticles against weathering of coated wood. To approach this purpose, the modified water based acrylic coating was applied on beech wood surface and then the treated wood specimens were exposed to natural weathering.

## 2 MATERIALS AND METHODS
## 2. MATERIJALI I METODE

Defect-free beech (*Fagus orientalis* L.) wood specimens (12 cm × 10 cm × 1 cm; L×T×R) were cut from air dried boards and then sanded using P220 sandpaper. The transparent water-based acrylic (methyl methacrylate-styrene copolymer) with 42 wt% solid content was obtained from Newcolour/Iran. Nano-sized ZnO particles (average size of 60 nm) and 2, 4 -dihydroxy benzophenone as UV stabilizers were provided by Alfa Aesar /USA and Sigma Alderich/ Germany, respectively. In this study, raw wood as a control sample (CS) unmodified (UN), modified with 0.5 wt% of dihydroxy benzophenone (UA) and 0.5 wt% of ZnO nano particles (ZN) coatings were applied on wooden surfaces by brush and allowed to dry for 48 hours. There was only one coating layer and its quantity was adjusted to approximately 120 g/m$^2$ for all samples. To modify the coating with dihydroxy benzophenone, it (0.42 g) was dissolved in ethanol (10 ml) and then mixed with an acrylic coating (200 g) using a magnetic stirrer for 15 min. To modify the coating with ZnO nanoparticles, 10 g of ZnO nanoparticles was suspended in distilled water (190 g) using sonication for 30 min. The suspensions (8.4 g) were then added to the coating (200 g) and mixed using a magnetic stirrer for 15 min.

### 2.1 Natural weathering
### 2.1. Prirodno izlaganje vremenskim utjecajima

The weatherability of wood specimens finished with modified coatings was assessed using natural weathering. In order to approach this purpose, the specimens were exposed outdoors facing south at an angle of 45° to the horizontal in Gorgan, Iran from March to September 2017.

### 2.2 Field emission scanning electron microscope (FE-SEM)
### 2.2. Pretražni elektronski mikroskop s emisijom polja (FE-SEM)

To observe and confirm the nano-scale size and morphology of ZnO nanoparticles, a FE-SEM (MRA3, TESCAN Co.) operating at voltage of 10 kV was used. The diameter of 100 ZnO nanoparticles was measured using DigiMizer software.

**Table 1** Average weather conditions in Gorgan from March to September 2017
**Tablica 1.** Prosječne vrijednosti vremenskih uvjeta za Gorgan od ožujka do rujna 2017.

| **Daily temperature, °C** *Dnevna temperatura,* °C | **Daily minimum temperature, °C** *Najniža dnevna temperatura,* °C | **Daily maximum temperature, °C** *Najviša dnevna temperatura,* °C | **Daily relative humidity, %** *Dnevna relativna vlažnost zraka, %* | **Daily minimum relative humidity, %** *Najmanja dnevna relativna vlažnost zraka,* % | **Daily maximum relative humidity, %** *Najveća dnevna relativna vlažnost zraka,* % | **Monthly rainfall, mm** *Mjesečne padaline,* mm | **Monthly hours of sunshine, hrs** *Sunčani sati u mjesecu,* h |
|---|---|---|---|---|---|---|---|
| 21.16 | 11.4 | 35.6 | 66.63 | 26.8 | 93.8 | 19.1 | 273.3 |

### 2.3 Adhesion test
### 2.3. Ispitivanje adhezije

The pull-off method was used to evaluate adhesion strength between the wood and coating. All measurements were carried out with a Positest pull-off Adhesion Tester (DeFelsko, USA) based on ASTM D4541. The aluminum dollies of 20 mm diameter were glued onto the surface of the coated wood with an epoxy resin (UHU Plus, Germany) and allowed to cure for 24 hrs at room conditions. Then, a slot was cut into the coating film around the dollies. Finally, a dolly was separated by applying a force perpendicular to the surface test.

### 2.4 Color measurement
### 2.4. Mjerenje boje

The colors of specimens in $L^*a^*b^*$ coordinates were measured using a colorimeter (Lovi bond S500, USA) according to ASTM D2244. The color difference ($\Delta E^*$) was determined for each sample by calculating from initial and final values using the following equation (Eq. 1):

$$\Delta E^* = \sqrt{\Delta L^{*2} + \Delta a^{*2} + \Delta b^{*2}} \qquad (1)$$

where $\Delta a^*$, $\Delta b^*$ and $\Delta L^*$ are the changes between the initial and final interval values.

### 2.5 Contact angle tests
### 2.5. Ispitivanje kontaktnog kuta

The contact angle tests were performed using PG-X Measuring Head (Switzerland) according to ASTM D5946 and were measured by a droplet of deionized water (3 µl). In this study, the contact angles of the sample surface were determined before and after weathering from zero to 10 seconds and were repeated with five drops for each sample.

### 2.6 ATR-FTIR analysis
### 2.6. ATR-FTIR analiza

ATR-FTIR spectroscopy (Vertex 80 spectrometer, Bruker Optics, Germany) was applied to study coated samples during weathering. For all samples, band intensities were normalized using the band at 1450 cm$^{-1}$, which exhibited a negligible change during weathering (Forsthuber *et al*., 2013).

### 2.7 Stereomicroscope analysis
### 2.7. Analiza stereomikroskopom

Stereomicroscope (Olympus, Japan) was used to investigate the surface morphology of coated wood specimens during weathering.

## 3 RESULTS AND DISCUSSION
## 3. REZULTATI I RASPRAVA

Figure 1 shows the FE-SEM of ZN, confirming all ZNs were in the range of nanoscale size and had almost uniform size. The average diameter of ZN obtained was 55 ±16 nm.

### 3.1 Adhesion strength
### 3.1. Adhezivna čvrstoća

The adhesion strength of acrylic paint, as indicated by force used to separate glued dolly from the surface test, is shown in Figure 2 for weathered and

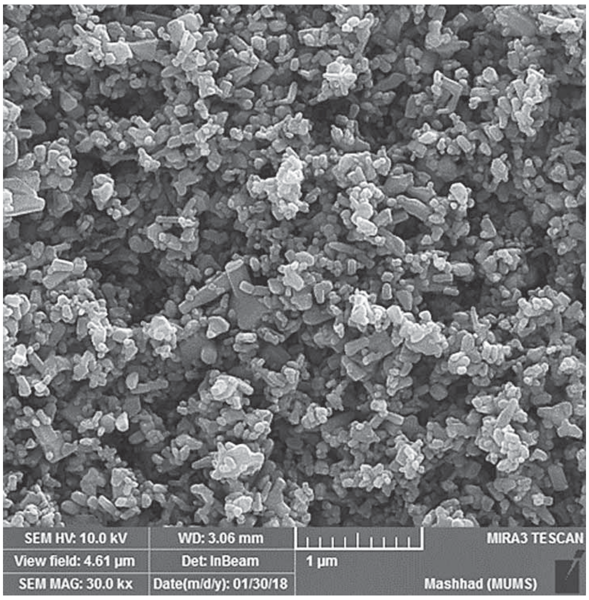

**Figure 1** FESEM of ZnO particles
**Slika 1.** FESEM ZnO čestica

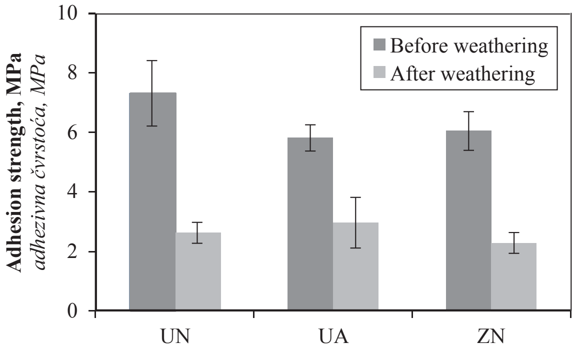

**Figure 2** Adhesion strength of samples before and after weathering
**Slika 2.** Adhezivna čvrstoća uzoraka prije i nakon izlaganja vremenskim utjecajima

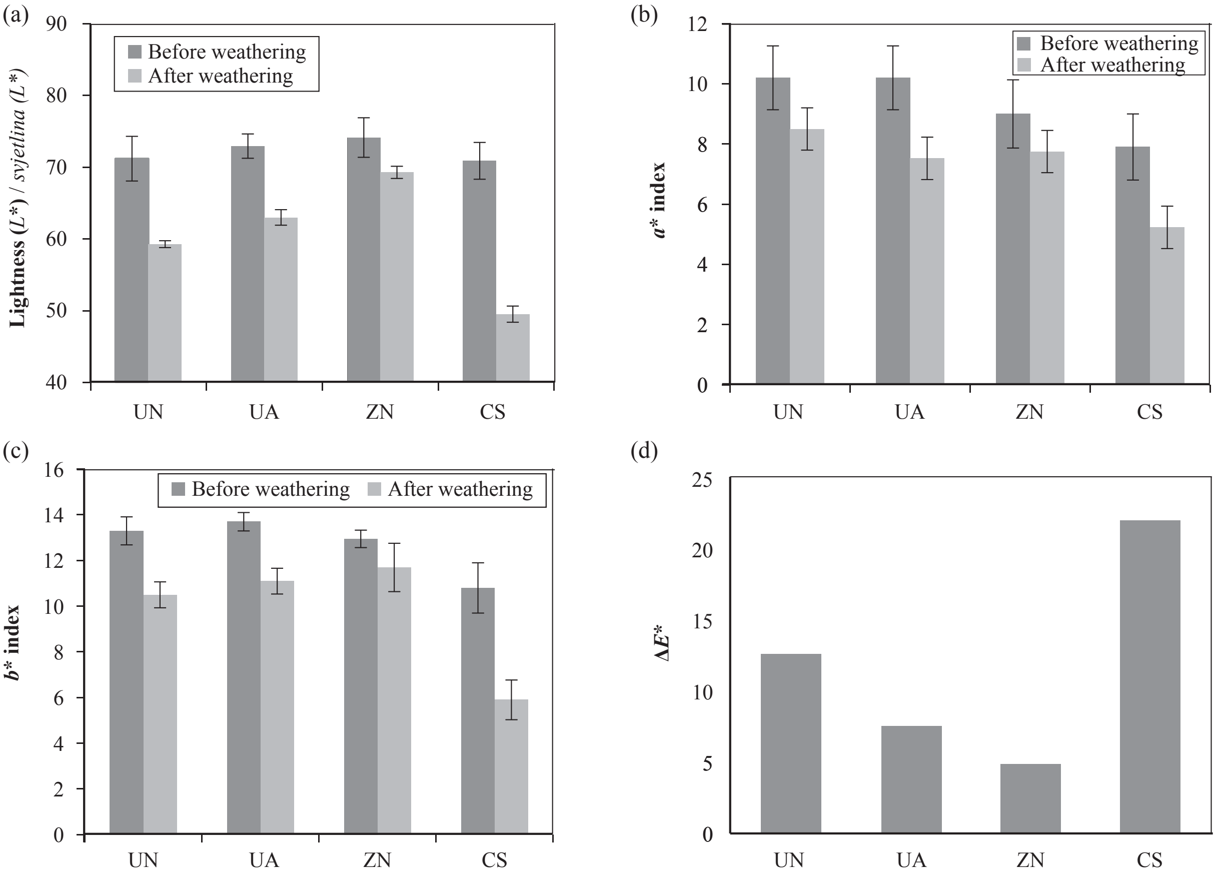

**Figure 3** Comparison of acrylic coatings stabilized with different additives: lightness (a), red–green index (b), yellow–blue index (c) and total color change (d) (Error bar shows standard deviation)
**Slika 3.** Usporedba akrilnih premaza stabiliziranih različitim aditivima: svjetlina (a), crveno-zeleni indeks (b), žuto-plavi indeks (c) i ukupna promjena boje (d) (traka pogreške prikazuje standardnu devijaciju)

unweathered specimens. In unweathered cases, unmodified coating (UN) showed the highest value, while modified coating containing ZnO and dihydroxy benzophenone (ZN and UA) showed a reduction in adhesion strength. This reduction has been previously reported in the presence of nano ZnO (Miklečić *et al*., 2017). Unfortunately, ZnO nanoparticles exhibit a high tendency to agglomerate in the polymer matrix because of its large surface area and high surface energy (Kathalewar *et al*., 2013). The results obviously revealed that six months of outdoor conditions significantly reduced the adhesion of coating. The loss of adhesion was previously reported for wood transparent coating during weathering (Singh and Dawson, 2003). Solar radiation penetrated through transparent coating on the surface of wood and degraded underlying wood, causing failure between wood and paint. These results confirmed that the coating modified with dihydroxy benzophenone and ZnO was more effective in reducing adhesion loss of applied coatings during weathering than unmodified coatings. It is interesting to note that the dominant failure types in the pull-off test were the adhesive failures between wood and coating.

### 3.2 Color measurement
### 3.2. Mjerenje boje

The results of color indexes (*L**, *a**, *b**) and color differences before and after six months of weathering are presented in Figure 3a. The results show that the weathering process was effective on lightness (*L**) in such a way that all samples became darker than those unweathered. The majority of UV is absorbed by lignin in wood and its photooxidation causes the formation of yellow to brownish compounds (ortho and para quinonoid compounds), resulting in the initial dark-brown color of wood during weathering. Additionally, the mold or mildew growth on the wood or coating surfaces usually appears as black spots during weathering (Nejad and Cooper, 2017). The highest change of *L** was observed in the case of UN and the lowest with ZN. For all

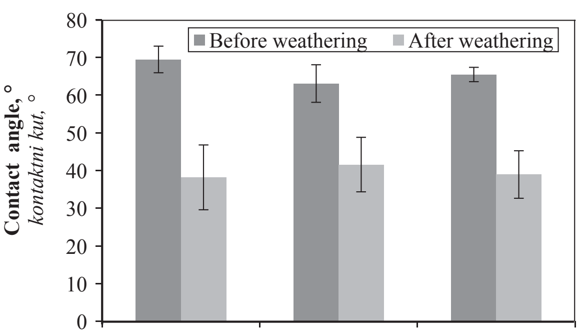

**Figure 4** Contact angle diagram for different UV absorbents before and after weathering (Error bar shows standard deviation)
**Slika 4.** Prikaz kontaktnog kuta za različite UV apsorbere prije i nakon izlaganja vremenskim utjecajima (traka pogreške prikazuje standardnu devijaciju)

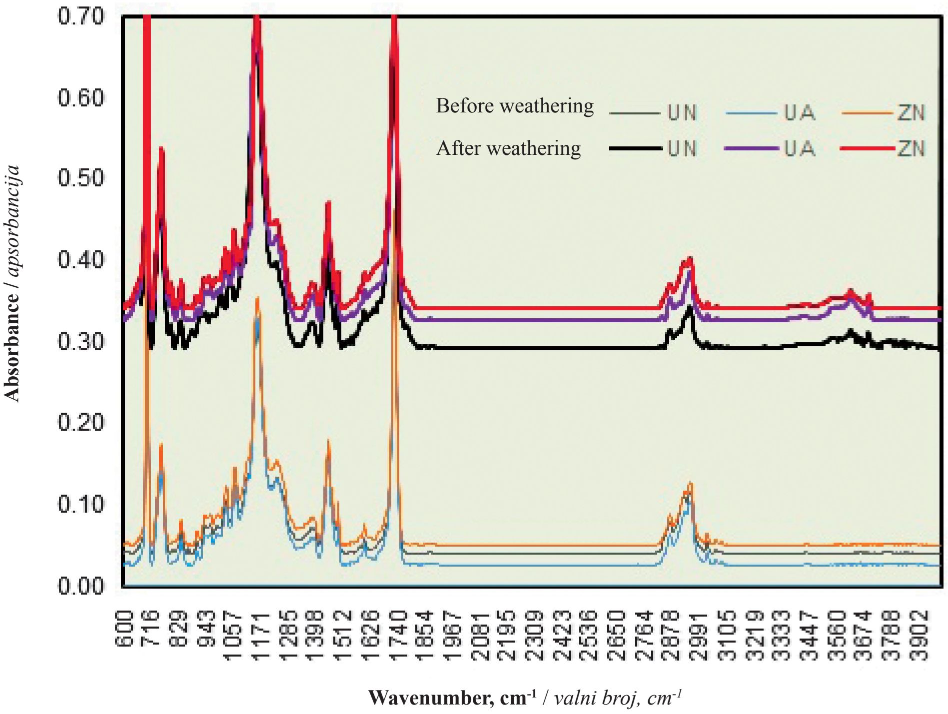

**Figure 5** Infrared spectra of coated samples containing different UV absorbents before and after natural weathering
**Slika 5.** Infracrveni spektar premazanih uzoraka koji sadržavaju različite UV apsorbere prije i nakon izlaganja vremenskim utjecajima

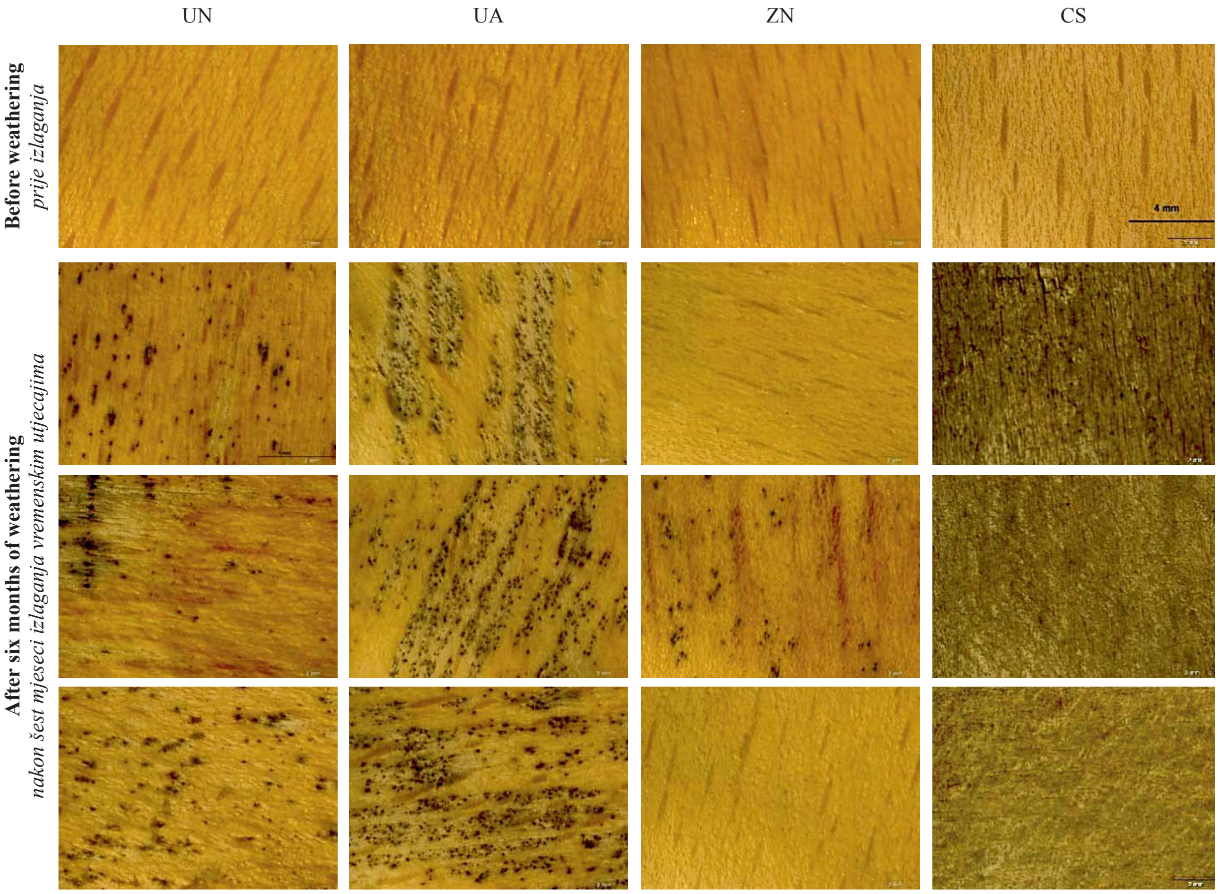

**Figure 6** Visual assessments of uncoated and coated beech surface before (one replicate) and after weathering (tree replicates)
**Slika 6.** Vizualna ocjena nepremazane i premazane površine bukovine prije (jedan uzorak) i nakon izlaganja vremenskim utjecajima (tri uzorka)

coated specimens, $a^*$ and $b^*$ indexes were almost equally reduced after weathering (Figure 3b and 3c). The overall color changes ($\Delta E^*$) after six-month natural weathering are presented in Figure 3d. For uncoated wooden samples, the color change was 22.08, which was higher than that of the coated samples. This finding shows that the application of acrylic coating was effective to avoid weathering degradation. Furthermore, modifying coating with ZnO had a positive impact on the color stability of wooden samples during weathering. Miklečić et al. concluded that the addition of ZnO nanoparticles into the polyacrylate coating resulted in higher color stability of thermally modified beech wood samples during natural weathering. The weathering is usually accompanied by the growth of dark-colored spores of mold fungi on the wood surface, which causes more color changes in the appearance (Tracton, 2006). The reduction of color changes due to the use of ZnO is related to fungicide properties of this material that is evident in stereo microscopic images.

### 3.3 Contact angles
### 3.3. Kontaktni kutovi

The surface hydrophilicity of the samples was determined by contact angle measurement (Figure 4). The results showed that the surface hydrophilicity increased after weathering, where it was extreme for uncoated samples. An increased wettability of weathered samples is related to changes in the surface chemical compositions and the increase of surface roughness caused by cracks (Matuana and Kamdem, 2002; Meiron *et al*., 2004). According to Figure 4, the modification of coating was not effective on the decreasing of contact angle changes of samples during weathering.

### 3.4 ATR-FTIR
### 3.4. ATR-FTIR

ATR-FTIR experiments were performed to study the chemical degradation of the coatings before and after weathering (Figure 5). The main changes are related to a wide band in the region 3500 - 3700 $cm^{-1}$. This band belongs to the stretching vibration of hydroxyl of water absorbed in the coating during the weathering process. Furthermore, there are no noticeable changes in intensity peaks around 1730 , 2960, 1150 and 880 $cm^{-1}$, where the signs of oxidation degradation due to weathering could be expected *(*Allen *et al*., 1997; Nguyen *et al*., 2016). This revealed that during six-month natural weathering, all coatings were resistant to photooxidation.

### 3.5 Stereomicroscope analysis
### 3.5. Analiza stereomikroskopom

Visual assessments of the uncoated and coated beech were evaluated by stereomicroscope (Figure 6). In uncoated samples, mold growth caused disfigurement of weathered samples. In addition to abiotic factors (sunlight, metal ion, etc.), biotic factors (molds, fungi, etc.) affect the color changes during natural weathering (Mohebby and Saei, 2015). Mold fungi can also grow on the painted wood and penetrate into the paint film, thereby colonizing in the interface between wood and paint (Gobakken and Westin, 2008). *Aureobasidium pullulans* is the most common mold observed on weathered wood (Rowell, 2012). It is very resistant to stress factors. It can withstand temperatures up to 80 °C and pH range from 1.9 - 10.1 and survive for a long time without moisture (Kutz, 2005). So, the use of the proper additive is necessary for the paint structure in outdoor applications. The images of stereomicroscope revealed that hydroxy benzophenone, as UV stabilizer, could not prevent the expansion and growth of mold, while ZN, due to antifungal properties, was effective against mold growth. These results correspond to the report of Terzi *et al*. (2016).

## 4 CONCLUSIONS
## 4. ZAKLJUČAK

The modification of acrylic paint with 2, 4 -dihydroxy benzophenone revealed that it had no effect against natural weathering, evidenced by observing adhesion strength, color and contact angle changes. During weathering, ZnO nanoparticles reduced the color changes and mold growth on coated wood samples by absorbing a wide range of UV radiation.

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# Influence of Fibre Length on Properties of Glass-Fibre Reinforced Bark Particleboards

## Utjecaj duljine vlakana na svojstva iverice od kore ojačane staklenim vlaknima

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**ABSTRACT** • *This study was carried out to assess the feasibility of glass fibres of various lengths (12 mm, 18 mm, 24 mm and 30 mm) as reinforcement on the mechanical performance of bark particleboards intended for thermal insulation. To evaluate their efficiency, the results of fibre reinforced particleboards at mass of 3wt% concentration were compared with plain bark based boards. Thermal, physical and mechanical properties (modulus of rupture, modulus of elasticity and internal bond) were determined on unreinforced and reinforced specimens. In general, the results of the thermal conductivity measurements indicated that the bark panels could potentially be used as feedstock for thermal insulation panels. However, the glass fibres lengths had a direct adverse effect on the mechanical behaviour of the bark particleboard, instead of providing synergistic reinforcement. Furthermore, the static bending properties, mainly the modulus of rupture, gradually decreased with increasing lengths of glass fibre.*

***Keywords:*** *utilization of bark; bio-based insulation panel; poplar bark*

**SAŽETAK** • *Istraživanje je provedeno kako bi se procijenila mogućnost uporabe staklenih vlakana različitih duljina (12; 18; 24 i 30 mm) kao materijala za ojačanje iverica proizvedenih od kore i namijenjenih toplinskoj izolaciji te kako bi se istražio utjecaj tih vlakana na mehanička svojstva iverica. Da bi se procijenio utjecaj staklenih vlakana, uspoređena su svojstva iverica ojačanih staklenim vlaknima masenog udjela vlakana od 3 % i iverica napravljenih od kore, bez ojačanja staklenim vlaknima. Uspoređena su toplinska, fizička i mehanička svojstva (modul loma, modul elastičnosti i međuslojna čvrstoća) uzoraka ploča iverica s ojačanjem staklenim vlaknima i bez ojačanja. U osnovi, rezultati mjerenja toplinske vodljivosti pokazali su da bi se ploče od kore mogle potencijalno upotrebljavati za toplinsku izolaciju. Međutim, duljina staklenih vlakana imala je izravan nepovoljan utjecaj na mehanička svojstva iverica od kore. Nadalje, svojstva statičkog savijanja, uglavnom modul loma, postupno su se smanjivala s povećanjem duljine staklenih vlakana.*

***Ključne riječi:*** *uporaba kore; izolacijske ploče na prirodnoj bazi; kora od topolovine*

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## 1 INTRODUCTION
## 1. UVOD

According to the European Union, the building sector is responsible for consuming 40 % of the total energy in Europe. Therefore, ways of improving thermal efficiency and decreasing greenhouse gases emissions, such as thermal insulation of building envelopes, is one of the most crucial methods to maximize the energy savings of a building during winter heating and summer air-conditioning (Schiavoni *et al*., 2016).

Bio-based thermal insulation materials cover a variety of natural fibre resources such as cellulose (Hurtado *et al*., 2016), bast fibres such as hemp and flax (Latif *et al*., 2014; Nguyen *et al*. 2016), cotton stalk (Zhou *et al*., 2010) coconut husk or sugarcane bagasse (Panyakaew and Fotios, 2011), rice straw (Wei *et al*., 2015), sunflower stalks (Mati-Baouche *et al*., 2014) or other crop by-products (Palumbo *et al*., 2015), wood shavings (Sekino, 2016), textile waste from plant fibres (Lacoste *et al*., 2018), and agglomerated cork (Barreca and Fichera, 2016) etc.

Bark (as a lignocellulosic residue), is mainly burned as a fuel for heat energy in wood mills. However, recent research efforts consider the use of bark as raw material in the production of particleboard (Yemele *et al*., 2008), wood-plastic composites (Yemele *et al*., 2010), and as thermal insulation material in the form of polyurethane foam (D'Souza *et al*., 2016) or rigid panels (Kain *et al*., 2016; Pásztory *et al*., 2017).

One of the essential limitations on the exploitation of bark particles in wood industry is the resulting weakness of the mechanical properties of the manufactured wood-based panels or composites. A potential solution to overcome this issue could be the reinforcement of bark particle boards with common synthetic fibres such as glass, carbon, basalt and aramid. Glass fibres were examined as reinforcing filaments in cement and concrete composites (Kizilkanat *et al*., 2015; Arslan, 2016) and wood-plastic composites (Zolfagari *et al*., 2015). The flexural strength and the compressive strength of the concrete can be improved by adding glass fibre to the matrix (Majumdar and Nurse, 1974; İskender and Karasu, 2018). The flexural properties of solid and laminated wood beams, plywood and particleboards were also investigated by overlaying glass fibre on the surface (Smulski and Ifju, 1987).

The objectives of this research were to i) examine the effect of the length of glass fibres on the thermal, physical and mechanical performance of glass fibres reinforced bark particleboards, ii) determine the most suitable critical length of glass fibres reinforcement and iii) investigate their market potential as thermal insulation panels.

## 2 MATERIALS AND METHODS
## 2. MATERIJALI I METODE

### 2.1 Materials
### 2.1. Materijali

The whole bark samples (inner and outer bark) used in this research were directly collected from the debarking units processing harvested poplar (*Populus euramericana cv.* Pannónia) logs with a wide diameter range, which were stored in a local sawmill in the area of Sopron, Hungary. The E-glass fibre roving used for this study was supplied by PD Tatneft-Alabuga Fiberglass LLC (Yelabuga, Russia) (Table 1). The lengths of 12 mm (GF_12), 18 mm (GF_18), 24 mm (GF_24) and 30 mm (GF_30) were manually cut from the fibreglass roving cylindrical packages (Figure 1) similarly as Tsalagkas *et al.* (2019). The main properties of the glass fibres and their lengths used in this work are given in Table 1 and Figure 1, respectively. The commercial UF resin and hardener used in this work was purchased from DUKOL Ostrava s.r.o. The resin properties at 20 °C were: solid content 67.65 %; pH value 8.8; dynamic viscosity 500 mPa·s; and Ford cup viscosity 84 seconds.

### 2.2 Particleboard manufacturing
### 2.2. Proizvodnja iverice

Initially, bark slices of various thicknesses (15-30 mm), consisting of inner and outer bark, were collected and dried below 20 % in a chamber. Consecutively, the inner and outer bark were cut into small pieces and

**Table 1** Main properties of glass fibres (data was provided by the manufacturer of glass fibres)
**Tablica 1.** Osnovna svojstva staklenih vlakana (podatci su dobiveni od proizvođača vlakana)

| **Product code** *Šifra proizvoda* | **Type of fibres** *Vrsta vlakana* | **Filament diameter, mm** *Promjer filamenta,* mm | **Linear density, tex** *Linearna gustoća,* tex | **MC**, % | **Breaking strength, gf/tex** *Prekidna čvrstoća,* gf/tex |
|---|---|---|---|---|---|
| EC 14-300-350 | E-glass<br>Silane modified | 14.0±1.5 | 300±15 | <0.20 | >45 |

**Figure 1** Glass fibres used in this study (a) roving, (b) fibres lengths and (c) mixed with bark particles
**Slika 1.** Staklena vlakna primijenjena u istraživanju: a) namotana, b) vlakna različite duljine, c) pomiješana s iverjem kore

chipped into particles using a hammer mill equipped with 8 mm screening holes. The granulated bark particles ranged from 0.5 mm to 8 mm fractions and were used as raw material for the manufacturing of bark panels. The moisture content of the bark particles was adjusted to 6 to 9 % before further processing. The randomly oriented, chopped glass fibres of the prepared length were placed in a laboratory blender and homogenized with the bark particles for five minutes, before pressing. A mass of 4 % urea formaldehyde (UF) adhesive was sprayed on and was mixed to the mixture of the bark particles and glass fibres.The adhesive contains 35 % of aqueous solution of ammonium sulphate $(NH_4)_2SO_4$ as catalyst. The glued mixtures were immediately manually layered and formed in a wooden frame into a mat. Thereafter, the frame was removed, and the mats were pre-pressed by hand to compact the materials without heat transfer.

Next, the mats were transferred to a single-opening hydraulic hot press machine (Siempelkamp). The pressing temperature was set at 180 °C with a pressing time of 18 seconds per millimetre of thickness. The initial press pressure was 2.86 MPa, which was reduced after 120 seconds to 2 MPa, and after 240 seconds to 1.15 MPa to decrease the risk of panel damage by vapour. Single-layered boards with dimensions of 500 mm × 500 mm × 20 mm with a target density of 350 kg/m$^3$ were produced.

### 2.3 Characterization
### 2.3. Karakterizacija

All the composite boards were kept at 20 °C and 65 % relative humidity, until equilibrium moisture content (EMC) was achieved, prior to experimental measurements. All these boards were cut and trimmed into various test specimens.

Apparent density $\rho$ at 9.5-10.2 % moisture content was measured on the same samples used for the mechanical tests, as the average of at least fifteen specimens. The mechanical bending test was done according to the standard (EN 323:1993). Dimensional stability of the specimens from thickness swelling (*TS*) and water absorption (*WA*) were calculated according to European Standard EN 317:1993.

Thermal conductivity was measured across the thickness of the composite boards (500 mm × 500 mm × 20 mm) with a heat flow meter using a guarded hotplate method. The thermal conductivity measuring equipment was designed and assembled at the Technical University of Budapest. The measurement started when the steady state was achieved, i.e. when the fluctuation of the last fifty per minute measurements was under 0.002 W/mK.

The bending strength (*MOR*) and modulus of elasticity (*MOE*) of bark composite boards were measured using a universal testing machine Instron 5506 (static three-point bending), in compliance with the applicable European Standard EN 310:1993 at a crosshead speed of 8 mm/min. The tensile strength perpendicular to the surface (internal bond, *IB*) was determined by using 50 mm × 50 mm specimens from each panel according to EN 319:1993, at a speed of 0.8 mm/min.

### 2.4 Statistical analysis
### 2.4. Statistička analiza

The analysis of variance (ANOVA) was applied using Statistica13 software (TIBCO Software Inc., USA) to statistically evaluate the influence of the length of glass fibres on the thermal, mechanical and dimensional stability of the glass fibre reinforced bark boards produced in the current study. All data were checked for normality (Shapiro–Wilk test) and homogeneity of variance (Levene's test), at a 5 % significance level. Extreme values that did not fit the homogeneity or normality were excluded from subsequent analyses. Post hoc tests were conducted with Tukey's HSD test method at 5 % significance level.

## 3 RESULTS AND DISCUSSION
## 3. REZULTATI I RASPRAVA

Table 2 presents the mean values of the physical, thermal and mechanical properties of the bark particleboards reinforced with various glass fibre lengths ranging from 12 mm to 30 mm with a 6 mm interval. The mean density of the unreinforced and glass fibre reinforced bark particleboards fluctuated from 373.21 to 387.57 kg/cm$^3$, while the moisture content was estimated between 9.58 % and 10.18 %. The dimensional stability (*TS %* and *WA %*) was found not to be statistically different among the boards, indicating that glass fibres had no influence on these properties. In addition, the thermal conductivity of the boards was shown to be similar, in the area of 0.074-0.078 W/m·K, among all the panels made of bark-glass fibre composite boards, whilst the thermal conductivity of the bark particleboards was calculated as 0.079 W/m·K. Compared with other bio-based natural insulation materials, the obtained values were higher or contiguous to other wood-based panels (Pásztory *et al*., 2017), yet in a similar range. It is evident that the addition of glass fibres, with their lengths varying from 12 mm up to 30 mm, had a negative effect instead of a synergistic one on the static bending properties of bark particleboards. The control boards had the highest *MOR* and *MOE* values compared to the reinforced bark boards. Further, the *MOR* and *MOE* decreased with increasing the fibre length from 12 mm to 30 mm. The boards reinforced with a glass fibre length of 12 mm indicated the best mechanical performance among the fibre lengths tested. For specimens with a 12 mm glass fibre length, the *MOR* of the panels decreased by 28.17 % compared to control specimens. Furthermore, the *MOR* of 18 mm length panels additionally decreased by 17.65 % (related to 12 mm), the *MOR* of 24 mm fibre length panels further decreased by 10.71 % (in relation to 18 mm) and the *MOR* of 30 mm glass fibres reinforced again decreased by 12.00 % compared to 24 mm long fibres. Accordingly, the *MOE* value gradually decreased from 0.24 GPa to 0.19 GPa with fibres 12 mm long and then remained constant with longer fibre lengths.

However, it has to be noted that difficulties in workability or flocculation of fibres during mixing were observed with increasing fibre lengths, at the suggested

**Table 2** Physical, thermal and mechanical properties of the proposed glass fibre reinforced bark particleboards. Numbers in brackets represent standard deviation values.
**Tablica 2.** Fizička, toplinska i mehanička svojstva uzoraka iverica od kore drva ojačanih staklenim vlaknima (brojevi u zagradama vrijednosti su standardne devijacije)

| **Physical properties** / *Fizička svojstva* | **Control** | **GF_12** | **GF_18** | **GF_24** | **GF_30** |
|---|---|---|---|---|---|
| $\rho$, kg/m$^3$ | 387.57<br>( 14.24) | 376.89<br>( 19.46) | 375.60<br>(14.82) | 377.63<br>(12.47) | 373.21<br>(15.80) |
| *MC*, % | 9.73<br>(± 0.39) | 9.66<br>(± 0.84) | 10.18<br>(± 0.09) | 9.86<br>(± 0.27) | 9.58<br>(± 0.28) |
| *WA*, % (24 h) | 182.47$^{a*}$<br>(25.73) | 193.23$^{a}$<br>( 28.0) | 173.87$^{a}$<br>(16.91) | 177.54$^{a}$<br>(18.29) | 190.94$^{a}$<br>(26.26) |
| *TS*, % (24 h) | 8.15$^{a}$<br>(0.99) | 8.88$^{a}$<br>( 1.12) | 9.18$^{a}$<br>(0.80) | 9.14$^{a}$<br>(1.15) | 9.28$^{a}$<br>(0.88) |
| **Thermal properties** / *Toplinska svojstva* | | | | | |
| $\lambda$, W/mK | 0.079<br>( 0.003) | 0.074<br>( 0.007) | 0.075<br>( 0.002) | 0.078<br>(0.004) | 0.076<br>(0.002) |
| **Mechanical properties** / *Mehanička svojstva* | | | | | |
| *MOR*, MPa | 1.42$^{c}$<br>(0.24) | 1.02$^{b}$<br>(0.21) | 0.84$^{a,b}$<br>( 0.18) | 0.75$^{a}$<br>( 0.22) | 0.66$^{a}$<br>( 0.19) |
| *MOE*, GPa | 0.24$^{c}$<br>(0.04) | 0.19$^{b}$<br>(0.03) | 0.15$^{a}$<br>(0.02) | 0.15$^{a}$<br>( 0.02) | 0.15$^{a}$<br>( 0.03) |
| *IB*, N/mm$^2$ | 0.11$^{a,b}$<br>(± 0.01) | 0.13$^{b}$<br>(± 0.02) | 0.10$^{a,b}$<br>(± 0.03) | 0.09$^{a}$<br>(± 0.01) | 0.12$^{a,b}$<br>(± 0.03) |

*Mean specimen values and standard deviations (expressed in brackets), obtained in different groups. The same letters indicate statistically insignificant differences ($p$>0.05).

** Srednje vrijednosti i standardne devijacije (prikazane u zagradama) dobivene u različitim skupinama. Ista slova znače da među označenim vrijednostima nema statistički značajne razlike (p>0,05).*

3wt% concentration. A possible explanation for the decreasing flexural rupture could be a combination of several factors, such as the shear stresses distribution, the air voids in the panel material and the overlapping degree of the reinforced bark particleboards with longer glass fibre lengths. Furthermore, statistically, the IB values were not significantly affected by the reinforcement, demonstrating that the glass fibres had no interaction with the urea-formaldehyde bonded bark particles, therefore indicating no cohesion between the glass fibres and bark particles. This assumption was made because the glass fibres area on the breaking surface appears in higher ratio than the ratio of glass fibres in the volume. Consequently, the connection between the fibres and the urea formaldehyde glued bark particles was weaker than the bonding between glued bark particles. The bonding connection between urea formaldehyde resin and glass fibre was investigated by Sharma *et al*. (2015) from different mechanical aspects, but the positive effect of the glass fibre was not apparent.

## 4 CONCLUSIONS
## 4. ZAKLJUČAK

Glass fibre reinforced bark particleboards were evaluated as thermal insulation panels. The thermal conductivity investigation demonstrated the viability of the proposed boards as insulation panels. However, test results showed a negative influence especially on the bending strength (*MOR*) of the reinforced bark particleboards using the various glass fibre lengths. As the glass fibre length became longer, the flexural strength of reinforced bark particleboards reduced significantly. Therefore, the glass fibre 12 mm long demonstrated the best performance among the tested specimens. In addition, the internal bond strength of the reinforced specimens was not statistically different compared to control bark particleboards, indicating no cohesion between the glass fibres and the bark particles bonded with urea formaldehyde adhesive. Therefore, this study will be focused on i) the use of shorter glass fibre lengths such as 6 mm and 3mm and ii) the improvement of interfacial interaction between the bark particles and glass fibres through surface modification methods, or by examining other adhesives.

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THERMODOMINUS THERMODUX THERMOREX

GALEKOVIĆ
*Kvaliteta u tradiciji*

**Tvornica parketa**

DUX: Gotovi lakirani masivni klasični parket
DOMINUS: Gotovi lakirani masivni klasični parket - širina 9 cm
REX: Gotovi masivni lakirani podovi - uljeni / lakirani
Termo tretirani podovi: THERMODUX, THERMODOMINUS, THERMOREX
Eksterijeri: fasade, decking

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# Drvo tinea (*Weinmannia trichosperma* Cav.)

## NAZIVI I PODRUČJE RASPROSTRANJENOSTI

*Weinmannia trichosperma* Cav. vrsta je drva iz porodice *Cunoniaceae*. Trgovački su nazivi te vrste: Indischer Apfelbaum (Njemačka); tineo (Velika Britanija); tinae (Čile); tarco (Argentina). Stabla *Weinmannia trichosperma* Cav. nalazimo u Južnoj Americi, u Čileu i Argentini. Tipično nalazište tinea su niže i srednje nadmorske visine.

## STABLO

Stablo doseže visinu od 20 do 30 m. Prsni je promjer debla 50 – 90 cm.

## DRVO

### Makroskopska obilježja

Srž drva je crvenkastosmeđa, s vidljvim mrljama, a bjeljika se bojom neznatno razlikuje od srži. Tekstura drva je fina do srednje gruba, jednolična, često s tamnim prugama i dekorativna, katkad kose žice. Drvo je rastresito porozno. Godovi drva su uski, s jasno vidljivom granicom među njima.

### Mikroskopska obilježja

Drvo je rastresito porozno. Traheje su sitne, brojne i pojedinačno raspoređene. Tile su oskudne. Drvni su traci srednje široki do široki.

### Fizička svojstva

| | |
|---|---|
| Gustoća apsolutno suhog drva, $\rho_o$ | 650...700 kg/m$^3$ |
| Gustoća prosušenog drva, $\rho_{12\text{-}15}$ | oko 710 kg/m$^3$ |
| Radijalno utezanje, $\beta_r$ | oko 4,0 % |
| Tangentno utezanje, $\beta_t$ | oko 8,0 % |
| Volumno utezanje, $\beta_v$ | oko 13,2 % |

### Mehanička svojstva

| | |
|---|---|
| Čvrstoća na tlak | oko 48 MPa |
| Čvrstoća na savijanje | oko 87 MPa |
| Modul elastičnosti | oko 10,8 GPa |

## TEHNOLOŠKA SVOJSTVA

### Obradivost

Drvo se dobro ručno i strojno obrađuje. Dobro se lijepi i površinski obrađuje.

### Sušenje

Drvo se dobro i polako suši. Sklonost promjeni oblika je malena.

### Trajnost i zaštita

Srž drva tinea neotporna je na gljive uzročnice truleži (razred otpornosti 4 – 5) i podložna je napadu termita (razred otpornosti S). Po trajnosti pripada razredu 5 te se može upotrebljavati isključivo u interijeru.

### Uporaba

Od drva tinea izrađuju se furniri, dijelovi namještaja, parket, tokareni drvni elementi, intarzije i rezbarije, a primjenjuje se i za uređenje interijera i eksterijera.

### Sirovina

Drvo postiže visoku cijenu. Na tržištu je dostupno u ograničenim količinama, u obliku trupaca manjih dimenzija i furnira.

### Napomena

Drvo *Weinmannia trichosperma* nije na popisu ugroženih vrsta međunarodne organizacije CITES, ali je na popisu ugroženih vrsta međunarodne organizacije IUCN. Vodi se kao vrsta drva najmanje zabrinjavajućeg opstanka. Riječ je o manje poznatoj i manje istraženoj vrsti drva.

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prof. dr. sc. Jelena Trajković
dr. sc. Iva Ištok

# Upute autorima

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## Upute

Predani radovi smiju sadržavati najviše 15 jednostrano pisanih A4 listova s dvostrukim proredom (30 redaka na stranici), uključujući i tablice, slike te popis literature, dodatke i ostale priloge. Dulje je članke preporučljivo podijeliti na dva ili više nastavaka. Tekst treba biti u *doc formatu*, u potpunosti napisan fontom *Times New Roman* (tekst, grafikoni i slike), normalnim stilom, bez dodatnog uređenja teksta.

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Posljednja stranica treba sadržavati titule, zanimanje, zvanje i adresu (svakog) autora, s naznakom osobe s kojom će Uredništvo biti u vezi.

Znanstveni i stručni radovi moraju biti sažeti i precizni. Osnovna poglavlja trebaju biti označena odgovarajućim podnaslovima. Napomene se ispisuju na dnu pripadajuće stranice, a obrojčavaju se susljedno. One koje se odnose na naslov označuju se zvjezdicom, a ostale uzdignutim arapskim brojkama. Napomene koje se odnose na tablice pišu se ispod tablica, a označavaju se uzdignutim malim pisanim slovima, abecednim redom.

Latinska imena trebaju biti pisana kosim slovima (italicom), a ako je cijeli tekst pisan kosim slovima, latinska imena trebaju biti podcrtana.

U uvodu treba definirati problem i, koliko je moguće, predočiti granice postojećih spoznaja, tako da se čitateljima koji se ne bave područjem o kojemu je riječ omogući razumijevanje ciljeva rada.

Materijal i metode trebaju biti što preciznije opisane da omoguće drugim znanstvenicima ponavljanje pokusa. Glavni eksperimentalni podaci trebaju biti dvojezično navedeni.

Rezultati trebaju obuhvatiti samo materijal koji se izravno odnosi na predmet. Obvezatna je primjena metričkog sustava. Preporučuje se upotreba SI jedinica. Rjeđe rabljene fizikalne vrijednosti, simboli i jedinice trebaju biti objašnjeni pri njihovu prvom spominjanju u tekstu. Za pisanje formula valja se koristiti Equation Editorom (programom za pisanje formula u MS Wordu). Jedinice se pišu normalnim (uspravnim) slovima, a fizikalni simboli i faktori kosima (italicom). Formule se susljedno obrojčavaju arapskim brojkama u zagradama, npr. (1) na kraju retka.

Broj slika mora biti ograničen samo na one koje su prijeko potrebne za objašnjenje teksta. Isti podaci ne smiju biti navedeni i u tablici i na slici. Slike i tablice trebaju biti zasebno obrojčane, arapskim brojkama, a u tekstu se na njih upućuje jasnim naznakama ("tablica 1" ili "slika 1"). Naslovi, zaglavlja, legende i sav ostali tekst u slikama i tablicama treba biti napisan hrvatskim i engleskim jezikom.

Slike je potrebno rasporediti na odgovarajuća mjesta u tekstu, trebaju biti izrađene u rezoluciji 600 dpi, crno-bijele (objavljivanje slika u koloru moguće je na zahtjev autora i uz posebno plaćanje), formata jpg ili tiff, potpune i jasno razumljive bez pozivanja na tekst priloga.

Svi grafikoni i tablice izrađuju se kao crno-bijeli prilozi (osim na zahtjev, uz plaćanje). Tablice i grafikoni trebaju biti na svojim mjestima u tekstu te originalnog formata u kojemu su izrađeni radi naknadnog ubacivanja hrvatskog prijevoda. Ako ne postoji mogućnost za to, potrebno je poslati originalne dokumente u formatu u kojemu su napravljeni (*excel* ili *statistica* format).

Naslovi slika i crteža ne pišu se velikim tiskanim slovima. Crteži i grafikoni trebaju odgovarati stilu časopisa (fontovima i izgledu). Slova i brojke moraju biti dovoljno veliki da budu lako čitljivi nakon smanjenja širine slike ili tablice. Fotomikrografije moraju imati naznaku uvećanja, poželjno u mikrometrima. Uvećanje može biti dodatno naznačeno na kraju naslova slike, npr. "uvećanje 7500 : l".

Diskusija i zaključak mogu, ako autori žele, biti spojeni u jedan odjeljak. U tom tekstu treba objasniti rezultate s obzirom na problem postavljen u uvodu i u odnosu prema odgovarajućim zapažanjima autora ili drugih istraživača. Valja izbjegavati ponavljanje podataka već iznesenih u odjeljku *Rezultati*. Mogu se razmotriti naznake za daljnja istraživanja ili primjenu. Ako su rezultati i diskusija spojeni u isti odjeljak, zaključke je nužno napisati izdvojeno. Zahvale se navode na kraju rukopisa. Odgovarajuću literaturu treba citirati u tekstu, i to prema harvardskom sustavu (*ime – godina*), npr. (Bađun, 1965). Nadalje, bibliografija mora biti navedena na kraju teksta, i to abecednim redom prezimena autora, s naslovima i potpunim navodima bibliografskih referenci. Popis literature mora biti selektivan, a svaka referenca na kraju mora imati naveden DOI broj, ako ga posjeduje (http://www.doi.org) (provjeriti na http://www.crossref.org).

## Primjeri navođenja literature

Članci u časopisima: Prezime autora, inicijal(i) osobnog imena, godina: Naslov. Naziv časopisa, godište (ev. broj): stranice (od – do). Doi broj.

Primjer

Kärki, T., 2001: Variation of wood density and shrinkage in European aspen (*Populus tremula*). Holz als Roh- und Werkstoff, 59: 79-84. http://dx.doi.org/10.1007/s001070050479.

Knjige: Prezime autora, inicijal(i) osobnog imena, godina: Naslov. (ev. izdavač/editor): izdanje (ev. svezak). Mjesto izdanja, izdavač (ev. stranice od – do).

Primjeri

Krpan, J., 1970: Tehnologija furnira i ploča. Drugo izdanje. Zagreb, Tehnička knjiga.

Wilson, J. W.; Wellwood, R. W., 1965: Intra-increment chemical properties of certain western Canadian coniferous species. U: W. A. Cote, Jr. (Ed.): Cellular Ultrastructure of Woody Plants. Syracuse, N.Y., Syracuse Univ. Press, pp. 551- 559.

Ostale publikacije (brošure, studije itd.)

Müller, D., 1977: Beitrag zür Klassifizierung asiatischer Baumarten. Mitteilung der Bundesforschungsanstalt für Forstund Holzvvirt schaft Hamburg, Nr. 98. Hamburg: M. Wiederbusch.

Web stranice

***1997: "Guide to Punctuation" (online), University of Sussex, www.informatics.sussex.ac.uk/department/docs/punctuation/node00.html. First published 1997 (pristupljeno 27. siječnja 2010).

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Examples:
Krpan, J. 1970: Tehnologija furnira i ploča. Drugo izdanje. Zagreb: Tehnička knjiga.
Wilson, J.W.; Wellwood, R.W. 1965: Intra-increment chemical properties of certain western Canadian coniferous species. U: W. A. Cote, Jr. (Ed.): Cellular Ultrastructure of Woody Plants. Syracuse, N.Y., Syracuse Univ. Press, pp. 551-559.

Other publications (brochures, studies, etc.):
Müller, D. 1977: Beitrag zur Klassifizierung asiatischer Baumarten. Mitteilung der Bundesforschungsanslalt für Forst- und Holzwirtschaft Hamburg, Nr. 98. Hamburg: M. Wiederbusch.

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***1997: "Guide to Punctuation" (online), University of Sussex, www.informatics.sussex.ac.uk/department/docs/punctuation/node00.html. First published 1997 (Accessed Jan. 27, 2010).

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HRVATSKA KOMORA
INŽENJERA ŠUMARSTVA
I DRVNE TEHNOLOGIJE

# HRVATSKA KOMORA INŽENJERA ŠUMARSTVA I DRVNE TEHNOLOGIJE

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