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Bonding Strength of Beech Plywood Glued with Alcohol-Soluble Phenol-Formaldehyde Resin

Čvrstoća lijepljenja bukove furnirske ploče fenol-formaldehidnom smolom topljivom u alkoholu

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ABSTRACT • *The present study examines the bonding strength of five-layer plywood made from beech wood (Fagus sylvatica L.). An alcohol-soluble phenol-formaldehyde (PF) resin as an adhesive between the veneer sheets was used. Various pressing factors, showing the effect of the varying ratio between them, were applied. The experimental samples both in a dry state and after immersion in boiling water for one hour were tested. The results clearly demonstrated that the bonding strength is very high when using the alcohol-soluble PF resin and meets the requirements of the standard BDS EN 314-2: 2002. The best bonding strength values were obtained at a press temperature of 145 °C, adhesive spread of 150 to 170 g/m², and initial adhesive temperature of 30-40 °C.*

KEYWORDS: *plywood; PF resin; bonding strength; Fagus sylvatica L.*

SAŽETAK • *U radu se ispituje čvrstoća lijepljenja peteroslojne furnirske ploče izrađene od bukovine (Fagus sylvatica L.). Za lijepljenje listova furnira upotrijebljena je fenol-formaldehidna (PF) smola topljiva u alkoholu. Proučavani su različiti parametri prešanja te njihovi međusobni odnosi. Ispitivani su suhi uzorci i uzorci nakon jednosatnog potapanja u kipućoj vodi. Rezultati su jasno pokazali da je čvrstoća lijepljenja furnirske ploče vrlo visoka kada se rabi PF smola topljiva u alkoholu te da zadovoljava zahtjeve standarda BDS EN 314-2: 2002. Najbolje vrijednosti čvrstoće lijepljenja dobivene su pri temperaturi prešanja od 145 °C, uz količinu nanosa ljepila 150 – 170 g/m² i uz početnu temperaturu ljepila 30 – 40 °C.*

KLJUČNE RIJEČI: *furnirska ploča, PF smola; čvrstoća lijepljenja; Fagus sylvatica L.*

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1 INTRODUCTION
1. UVOD

Plywood is a composite material mainly applied in the furniture and construction industries. It consists of several thin layers of veneer tightly glued together with an odd number of layers with the grain direction of adjacent layers oriented perpendicular to one another.

Obtaining quality adhesive joints between veneer sheets in the production of laminated wood and more specifically plywood is directly dependent on the main process factors. These factors can be briefly considered as factors characterizing the veneer and plywood package; factors characterizing the properties and use of the adhesive; and factors characterizing the conditions of pressing (Hong and Park, 2017).

The type of wood used directly affects the performance indicators of the finished plywood. In Bulgaria, various types of wood for plywood production are applied, but the most commonly used are beech (*Fagus sylvatica* L.) and poplar (*Populus* sp.). The reason for this is that beech wood has very good physical and mechanical properties (Gryc *et al.*, 2008; Rais *et al.*, 2022). In addition, this wood species is present in relatively large quantities in our country (approximately 144 million m^3). The distribution of beech is approximately 20 % of the total stock of the country (Executive Forest Agency, 2020).

In the process of developing logs and obtaining veneer sheets, roughness has a significant impact on the bonding process (Dundar *et al.,* 2008; Bekhta *et al.,* 2009). Micro and macro irregularities on the veneer surface are formed. Studies show that the strength of the adhesive layer increases with increasing roughness of the veneer, but up to certain values, after which a significant decrease is observed (Aydin, 2004; Wang *et al.,* 2006). The moisture content of the veneer should be in the range of 6 to 12 % (Aydin *et al.*, 2006; Quiao, 2014), and when bonding with multi-component adhesives, it should not be higher than 7 %. The investigations regarding the moisture content of the veneer show that at very low values, the adhesive viscosity significantly increases because of intensive moisture absorption from the applied adhesive (Resnik and Šernek, 2000). The viscosity of the adhesive decreases in cases of very high moisture content of the veneer. Both cases are a prerequisite for creating internal stresses, leading to reduction in the quality of the bonding joints. The piezo-thermal treatment process creates conditions for an increased amount of steam-gas mixture at high moisture content in the veneer. This can contribute to thermal degradation of the resin (Resnik and Šernek, 2000; Bekhta *et al.,* 2012; Bekhta *et al.,* 2020).

The initial temperature of the wood also has an influence on the quality of adhesive bonds. The viscosity of the resin decreases at higher temperatures of the wood due to the increased thermal movement of molecules. This, in turn, is a prerequisite for better adhesive bonds and penetration of the adhesive into the veneer sheets during the pressing process of the plywood package (Frihart, 2012; Tran *et al.,* 2020). The initial temperature of the adhesive affects the improvement of the contact between the adhesive and the wood, thus creating conditions for speeding up the curing process (Demirkir *et al.,* 2017; Bliem *et al.,* 2019; Sutiawan *et al.,* 2022).

The adhesive consumption and its optimal thickness are essential both practically and technologically (Apsari and Tanaka, 2023). The curing time of the resin in the adhesive bond is crucial. Insufficient pressing time results in the defect of weak bonding. Excessive pressure and temperature can lead to the destruction of the already cured resin, resulting in poor adhesive bond, and hence a decrease in the quality of the finished plywood (Kurowska *et al.,* 2010; Li *et al.,* 2020; Wei *et al.,* 2021).

The condition of the adhesive is primarily determined by its concentration, viscosity, and temperature (Gomez-Bues and Haupt, 2010). Phenol formaldehyde (PF) resins are widely used in the manufacture of construction plywood and oriented strand board (OSB) for exterior applications. PF adhesives are characterized with excellent bonding strength, water resistance, bioresistance and weather durability (Gomez-Bueso and Haupt 2010; Karthäuser *et al.,* 2024). Depending on the conditions under which the reaction is carried out, two types of resins can be obtained, resole (thermosetting) and novolac (thermoplastic). Resole resins are obtained by condensing phenol with an excess of formaldehyde, in the presence of a basic catalyst. They have the ability to transform into an insoluble and unmeltable state upon heating, as they undergo three stages of structural change – resole, resitol, and resite (Figure 1).

The high content of polar groups (hydroxyl and methylol) provides excellent adhesion of resol resins to wood, as well as their solubility in alcohol, bases, and water. The chain molecules of resols consist of phenolic nuclei linked together by methylene groups or ether bonds. Novolac resins are obtained in excess of phenol in the presence of an acidic catalyst. Upon heating, these resins cannot be converted into an unmeltable and insoluble state as easily as resol resins. The rapid curing of novolac resins only occurs in the presence of special curing agents (Sarika *et al.,* 2020).

Alcohol-soluble PF resins are of interest, because the glued products are characterized by high strength and resistance to external influences (Lee *et al.,* 2014; Popovska *et al.,* 2014; Shishlov *et al.,* 2015; Chi and Trang, 2021). The literature data show that research in this area is scarce. Therefore, the aim of the present study was to examine the properties of this adhesive under different bonding technological factors, as well as its influence on the quality characteristics of ply-

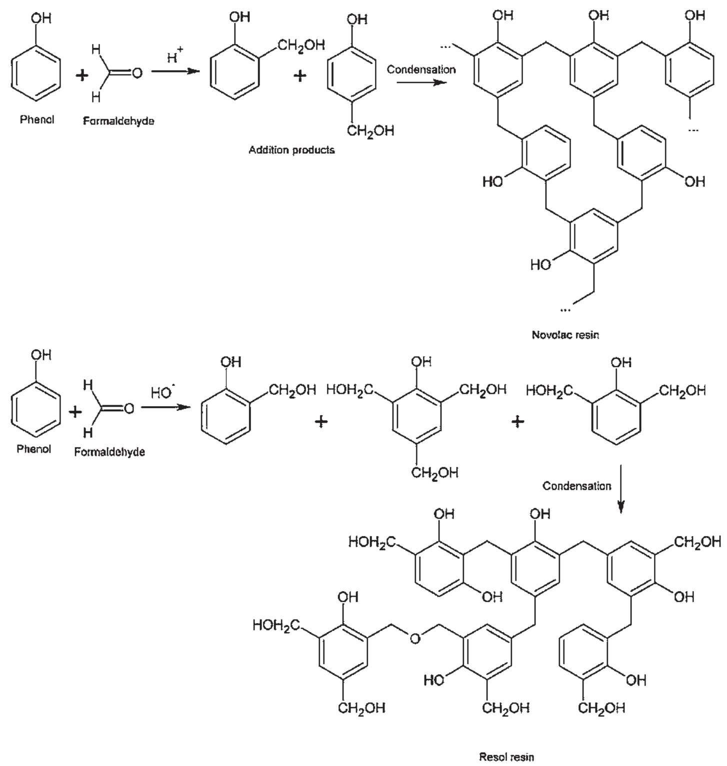

Figure 1 Synthesis of novolac and resol resin
Slika 1. Sinteza novolaka i rezol smole

wood products (in dry condition and after immersion of the test specimens in boiling water for one hour).

2 MATERIALS AND METHODS
2. MATERIJALI I METODE

The experimental studies in the present work were performed using high-quality beech veneer (*Fagus sylvatica* L.) with a thickness of 1.5 mm, free from wood defects, with a smooth surface and without processing defects. The veneer was obtained and dried to a moisture content of 6-8 % under production conditions (Cooperation "Obnova", Cherni Osam village). A total of 12 pieces of laboratory-glued five-layer plywood were produced in order to establish the strength indicators of the adhesive joints between the veneer sheets. After piezo-thermal treatment, the panels were removed from the hot press and arranged in tight formations for 24 hours. Before testing the strength parameters of the adhesive joints, the plywood panels were conditioned for seven days at a temperature of 20 °C and an air humidity of 65 %.

Thirty specimens were cut from each plywood sheet, half of which were tested in a dry condition and the other half in boiling water for 1 hour. The alcohol-soluble PF resin Prefere 14J350 (Prefere Resins Austria GmbH) with a solids content of 46 %, viscosity- 372 MPas at 20 °C, and pH- 6-8, was used as an adhesive.

The research was conducted in the laboratory of wood-based panels at the University of Forestry, Sofia according to the standards BDS EN 314-1:2006 and BDS EN 314-2:2002. The five-layer plywood samples were made in a hydraulic press with dimensions of 600/600 mm. The adhesive was applied to the veneer sheets using a roller applicator. This study presents the influence of certain technological factors on the bonding strength. Test specimens for determining the bonding strength of plywood were prepared according to BDS EN 315:2002.

The bonding strength at the farthest adhesive joints from the face layers, namely the second and third adhesive joints, was established. The strength characteristics of the laboratory-produced veneer sheets of beech wood was determined according to Eq. 1.

$$f_v = \frac{F}{l_1 \cdot b_1} \tag{1}$$

Where: f_v – bonding strength, N/mm^2;
F – breaking force of test specimens, N;
l_1– bonding area length, mm;
b_1– bonding area width, mm.

The dimensions of the test specimens were determined as the arithmetic mean values of two measurements in the middle and their length. The experiments were performed on a universal testing machine with an accuracy of 0.1 N under the following conditions: in dry state at a moisture content of plywood not exceeding 8 %, and after immersion of the test specimens in boiling water for one hour. After testing the specimens, it was recorded whether the failure occurred at the adhesive joint or not. This is an additional indicator for characterizing the quality of the adhesive bond.

The influence of bonding duration on the bonding strength of plywood was investigated at three different press temperatures (135 °C, 145 °C, and 155 °C), an adhesive spread of 130 g/m^2 and a pressure of 1.8 MPa. The effect of adhesive consumption on bonding strength was also established at the following parameters: pressing pressure of 1.8 MPa, pressing temperature of 145 °C, pressing time of 6.15 minutes, and different adhesive consumption ranging from 110 to 190 g/m^2 (Pipíška *et al.,* 2023).

Studies related to the effect of the adhesive initial temperature at the moment of its application on the bonding strength of plywood were performed. Under all the above conditions, a glue spread of 130 g/m^2 was applied.

The obtained values were processed by the method of variational statistics, and the values between two samples were processed using the T-test.

3 RESULTS AND DISCUSSION
3. REZULTATI I RASPRAVA

In order to establish the influence of the pressing time on the bonding strength of plywood, experiments with different pressing time and at different temperatures (135 °C, 145 °C and 155 °C) were conducted. A glue spread of 130 g/m^2 and a pressing pressure of 1.8 MPa were applied.

The relationship between the press temperature and the pressing time on the bonding strength in a dry state at 135 °C is graphically illustrated in Figure 2a.

The results obtained showed a significant increase in bonding strength of 27 % in the range of 4:30 – 5:30 min. At this temperature, the highest bonding strength was found at a pressing time of 8:30 min. Lowering the pressing time in all cases led to a decrease in bonding strength.

The results of the conducted T-test showed statistical significance when comparing bonding strength at different pressing times. In the first case, from 4:30 to 5:30 min., p-value = 0.00007; between 5:30 and 6:30 min, p-value = 0.00015. The results were not statistically significant in the range of 6:30 to 7:30 min (p-value = 0.41110). In the last mode range from 7:30 to 8:30 min, the results displayed statistical significance again (p-value = 0.00039).

The presented results indicate that the bonding strength significantly decreased after immersion of the test specimens in boiling water for 1 hour (Figure 2b).

The lowest values were found at press temperature of 135 °C and pressing time of 4:30 minutes (1.00 N/mm^2), and the highest values (1.58 N/mm^2) when the pressing time reached 8:30 minutes. In this case, as well as in the case of testing in a dry state, the same dependence was observed, namely that with an increase in the pressing time, the bonding strength also increased. The enhancement of the adhesive joint strength with increasing pressing time was in the range of 8 to 18 %.

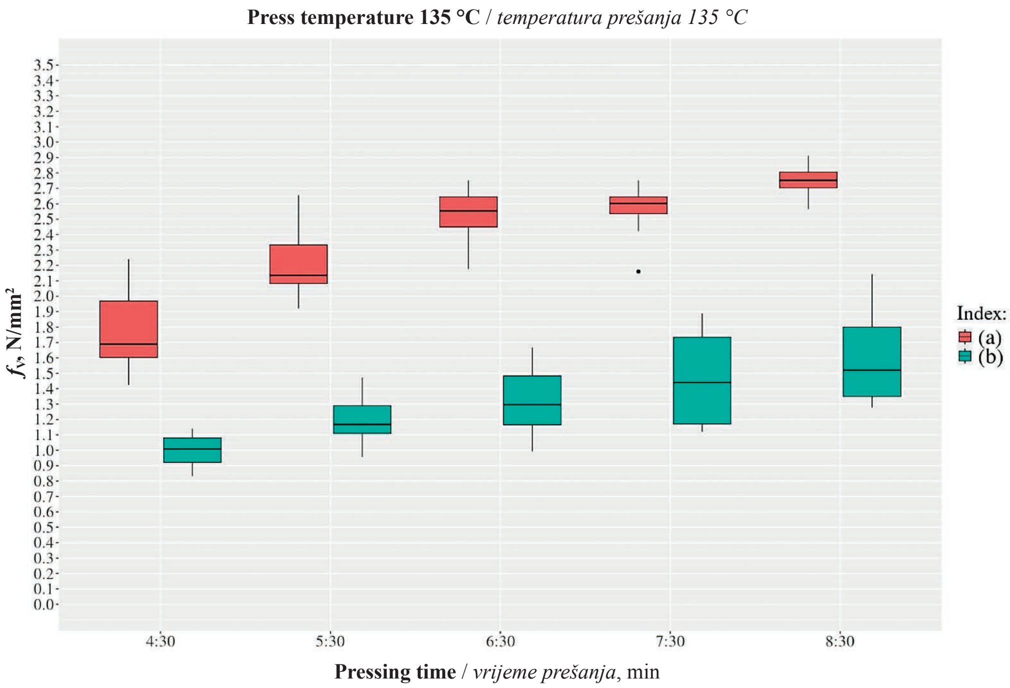

Figure 2 Influence of pressing time on bonding strength in a dry state (a) and after immersion of test specimens in boiling water for 1 hour (b) at press temperature of 135 °C
Slika 2. Utjecaj vremena prešanja na čvrstoću lijepljenja furnirske ploče u suhom stanju (a) i nakon jednosatnog potapanja uzoraka u kipućoj vodi (b) pri temperaturi prešanja od 135 °C

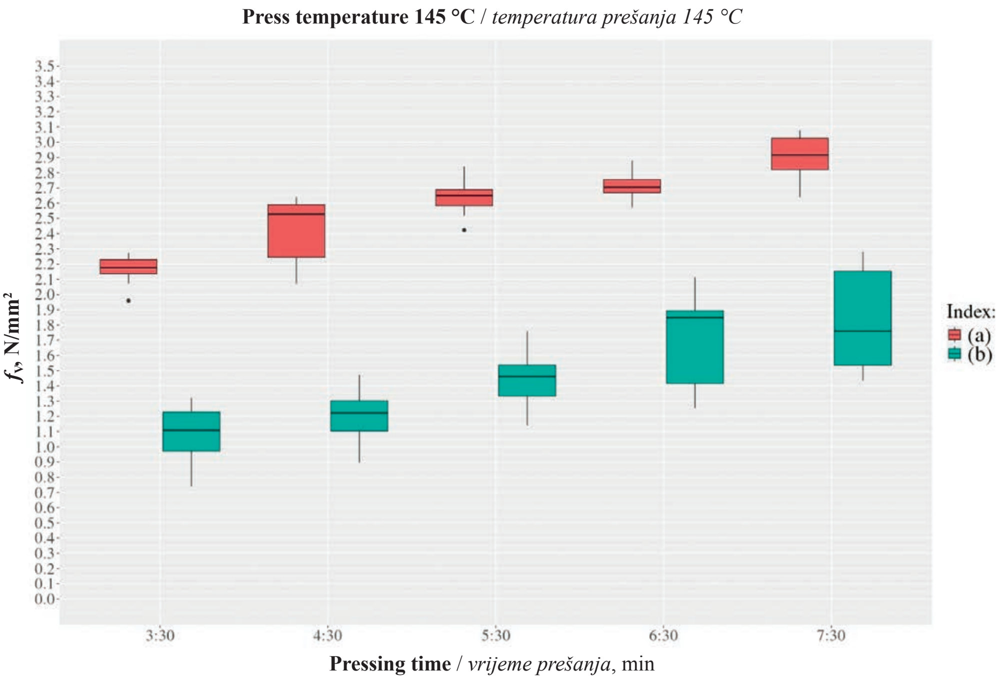

Figure 3 Influence of pressing time on bonding strength in a dry state (a) and after immersion of test specimens in boiling water for 1 hour (b) at press temperature of 145 °C
Slika 3. Utjecaj vremena prešanja na čvrstoću lijepljenja furnirske ploče u suhom stanju (a) i nakon jednosatnog potapanja uzoraka u kipućoj vodi (b) pri temperaturi prešanja od 145 °C

Increasing the press temperature from 135 °C to 145 °C in a dry state led to rising of bonding strength from 5 to 24 % (Figure 3a).

In this case, with increasing pressing time, the bonding strength rises from 6 to 11 %. The lowest bonding strength was observed at 3:30 min. (2.17 N/mm^2), while the highest value was found at 7:30 min (2.91 N/mm^2).

The results of the T-test showed the same dependence. In the first case, from 3:30 to 4:30 min, the p-value was 0.00026 and between 4:30 and 5:30 min, the p-value was 0.00101. In the range of 5:30 to 6:30 min, the p-value was 0.04331. In the last regime mode from 6:30 to 7:30 min, the results of the T-test were very small, which again shows statistical significance.

The results found at press temperature of 145 °C and immersion of the test specimens in boiling water for one hour showed similar dependencies to the previous cases (Figure 3b).

The lowest value was obtained with the shortest pressing time of 3:30 min. (1.09 N/mm^2), while the highest value was achieved with a pressing time of 7:30 min. (1.82 N/mm^2). The results clearly indicate that with rising pressing time in the specified range, the strength properties of the adhesive joints after a 1-hour stay in boiling water increased by approximately 67 %.

Increasing the temperature to 155 °C in a dry state exhibited the same dependence as in the other two cases (Figure 4a).

The most significant increase in the bonding strength was observed in the range between 5:30 and 6:30 min (approximately 9 %).

The values obtained from the performed T-test showed the same trend as in the previous cases. In the range from 2:30 to 3:30 min, p-value = 0.00034 and between 3:30 and 4:30 min, p-value = 0.00018. The values were negligibly small (significantly ≤0.05) from 4:30 to 6:30 min, which again showed statistical significance.

The action of boiling water influenced a negative effect on the bonding strength at a temperature of 155 °C (Figure 4b).

It increased to the greatest extent in the interval of 4:30-5:30 min, and the highest value (3.25 N/mm^2) was found at a pressing time of 6:30 min. The results of testing the experimental specimens, both in a dry state and after immersion in boiling water for 1 hour, showed that with increasing the press temperature and the pressing time, the bonding strength significantly enhanced. Pressing at higher temperatures results in higher bonding strength. In addition, a shorter pressing time at higher temperatures was used.

The results clearly show that this does not have a negative impact on the bonding strength of plywood glued with alcohol-soluble PF resin. This in turn is a prerequisite for intensifying the bonding process, which is an opportunity to increase the economic effect.

When determining the bonding strength in a dry state, the failure of the adhesive joints was from 75 to 90 % in the wood zone, due to the significantly large adhesion bonds. Another part of the specimens failed 100 % in the wood zone (8:30 min in Figure 2 as well as 7:30 min in Figure 3 and 6:30 min in Figure 4). The failure of the adhesive joints in the bonding strength test after im-

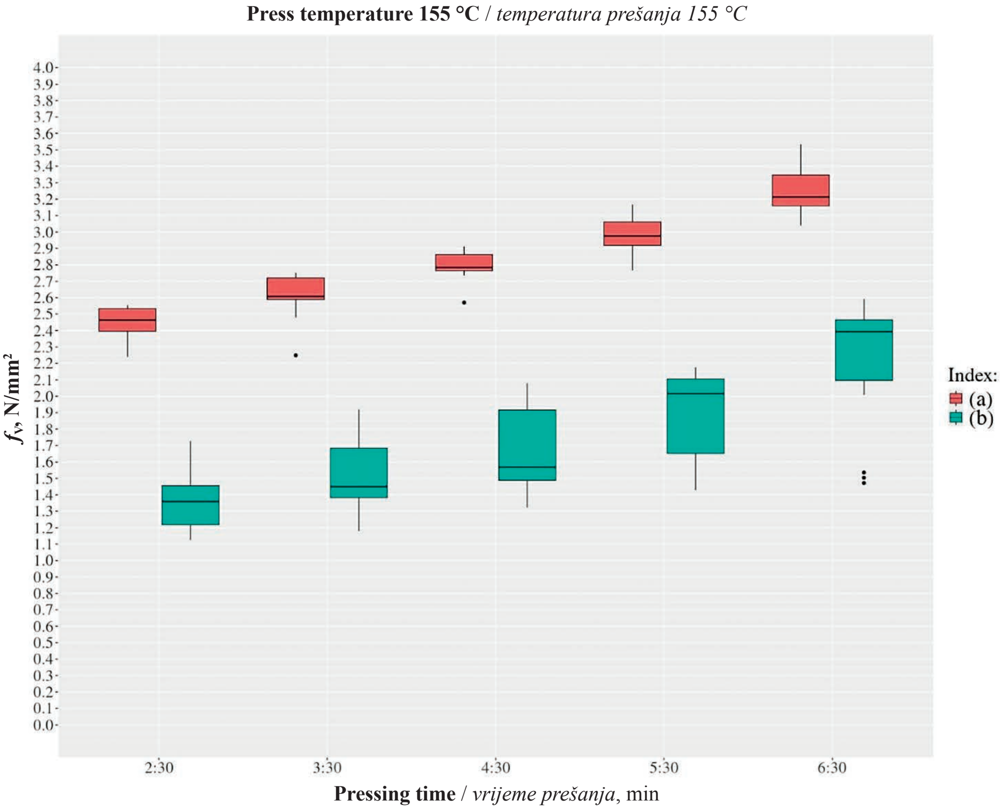

Figure 4 Influence of pressing time on bonding strength in a dry state (a) and after immersion of test specimens in boiling water for 1 hour (b) at press temperature of 155 °C
Slika 4. Utjecaj vremena prešanja na čvrstoću lijepljenja furnirske ploče u suhom stanju (a) i nakon jednosatnog potapanja uzoraka u kipućoj vodi (b) pri temperaturi prešanja od 155 °C

mersion of the test specimens in boiling water for 1 hour occurred from 10 to 40 % in the wood zone.

The obtained results were compared with the research of some authors in studies conducted in a similar area. Pipiška *et al*. (2024) applied different regimes, examining three types of specimens obtained under the following conditions: after conditioning at a temperature of 20 °C and relative humidity of 65 %; at soaking in water at 20 °C for 24 h; at boiling in water for 6 h and cooling in water at 20 °C at least 1 h. The determined shear strength of the adhesive joint was in the range of 1.1 to 1.82 N/mm² and the wood failure was from 33 to 85 %.

Other authors, such as Kallakas *et al*. (2020) considered different combinations of hardwood species (gray alder, black alder and aspen), the aim being to replace birch veneer in plywood. A consumption of PF adhesive of 152 to 179 g/m² was applied. The highest bonding strength value obtained was 2.39 N/mm², and the failure in the wood zone ranged from 43.3 to 79.8 % depending on the different processing parameters and combinations of wood species.

Interesting dependencies were found by Bekhta *et al*. (2020), who conducted a study on the bonding of birch plywood with phenol-formaldehyde resin at different pressing parameters and binder content. The shear strength found, depending on the applied conditions and adhesive consumption, was in the range of 1.2 to 2.93 N/mm². Other authors optimized some of the pressing processes when gluing birch plywood with phenol-formaldehyde resin. They investigated bond strength after soaking the test specimens at 20 °C for a period of 24 hours, as well as after boiling them in water for 4 hours. The established bond strength, depending on the selected pressing parameter, was in the range from 1.1 to 2.5 N/mm² (Kawalerczyk *et al*., 2022).

The above cases clearly show that not only the main pressing parameters but also the amount and type of the binder have a significant influence on the bonding strength of plywood.

The choice of optimal adhesive consumption for bonding plywood is of great importance. The adhesive consumption affects the cost of the final product. The influence of adhesive consumption on the bonding strength of plywood was determined by the following technological factors: pressing temperature of 145 °C; pressing time of 5:30 min; pressing pressure of 1.8 MPa. Glue spreading of 110, 130, 150, 170, and 190 g/m² was selected.

The results demonstrated that with increasing consumption of phenol-formaldehyde resin, the bonding strength in dry condition also increased. This was valid, however, only in the range of 110-170 g/m² (Figure 5).

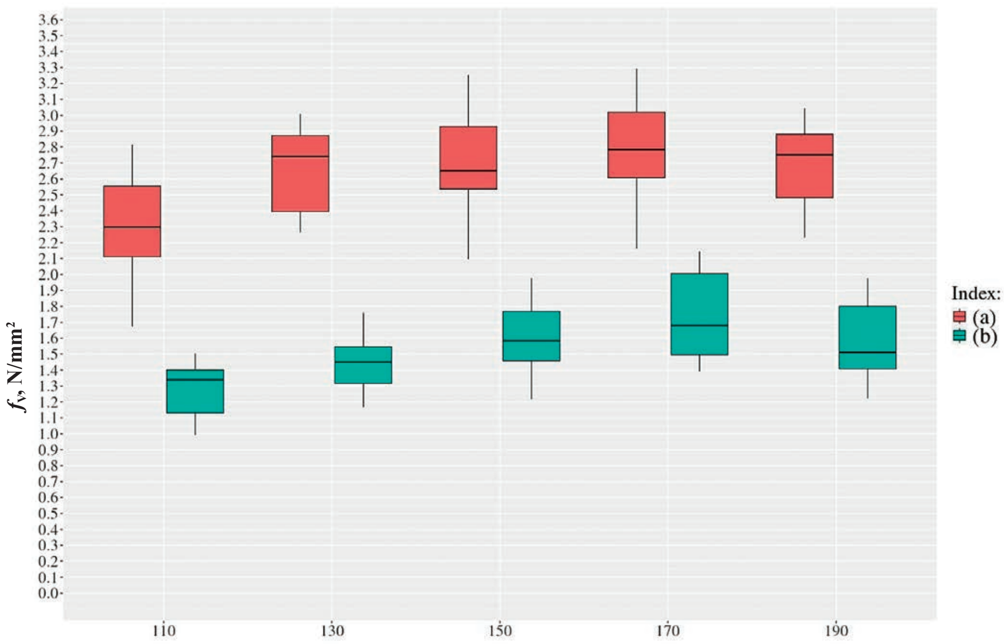

Figure 5 Influence of adhesive spread on bonding strength of plywood in a dry state (a) and after immersion in boiling water for one hour (b)
Slika 5. Utjecaj količine nanosa ljepila na čvrstoću lijepljenja furnirske ploče u suhom stanju (a) i nakon jednosatnog potapanja u kipućoj vodi (b)

A significant rise in bonding strength of about 15 % was observed when the adhesive consumption was increased from 110 to 130 g/m^2.

The bonding strength enhanced by about 3 % from 130 to 150 g/m^2. This dependency was repeated in the range of 150 to 170 g/m^2. When the adhesive spread reached 190 g/m^2, a decrease in bonding strength of plywood was observed. This is due to the formation of a thick adhesive joint, leading to significant stress within it. These results indicated that excessive increase in adhesive consumption is not advisable.

The results of the conducted T-test showed that there is no statistically significant difference in bonding strength when comparing different adhesive amounts. In the range of 110 to 130 g/m^2, p-value = 0.00644; from 130 to 150 g/m^2, p-value = 0.53025. Similar values were found for adhesive spread between 150 and 170 g/m^2 (p-value = 0.46423) and between 170 and 190 g/m^2 (p-value = 0.23646).

Testing of the specimens after immersion in boiling water for one hour showed a similar dependence as in the dry state, with the difference that the values were lower (Figure 5b).

In this case, there was again a sharp increase of about 13 % in the bonding strength from 110 to 130 g/m^2.

From the above, it becomes clear that the optimal consumption of alcohol-soluble phenol-formaldehyde resin can be assumed to be in the range of 150-170 g/m^2.

The initial temperature of the adhesive also influenced the bonding strength. The same process factors were applied as in the study of the adhesive consumption. When testing the samples in a dry state, the lowest bonding strength was obtained at an adhesive temperature of 10 °C – 2.02 N/mm^2 (Figure 6a).

As the adhesive temperature rises, the bonding strength also increases. It changed insignificantly (about 7 %) in the range of 10 to 20 °C. Further increase in temperature leads to a considerable enhancement in bonding strength (about 19 %) in the range of 20 to 30 °C. The increase of 19 % was also observed from 30 to 40 °C. The graphical representation shows that as the temperature rises in the range of 20 to 40 °C, the bonding strength of plywood increases from 2.26 to 3.32 N/mm^2, or by approximately 42 %.

Figure 6b illustrates the results of testing the samples after immersion in boiling water for one hour. A similar dependence was found as in the dry state, with the most intensive increase in bonding strength in the range of 30-40 °C. However, in this case, the values were lower compared to those in the dry state. The best bonding strength was achieved at the highest temperature.

The results of the conducted T-test showed that when comparing the bonding strength at different temperatures of the adhesive, in most cases statistical significance was assessed. When increasing the temperature from 10 to 20 °C, the results were not statistically significant (p-value = 0.10001). In a temperature range from 20 to 30 °C, p-value = 0.00019. In the final variant of 30 to 40 °C, p-value = 0.00053. The performed studies demonstrate that the optimal temperature of

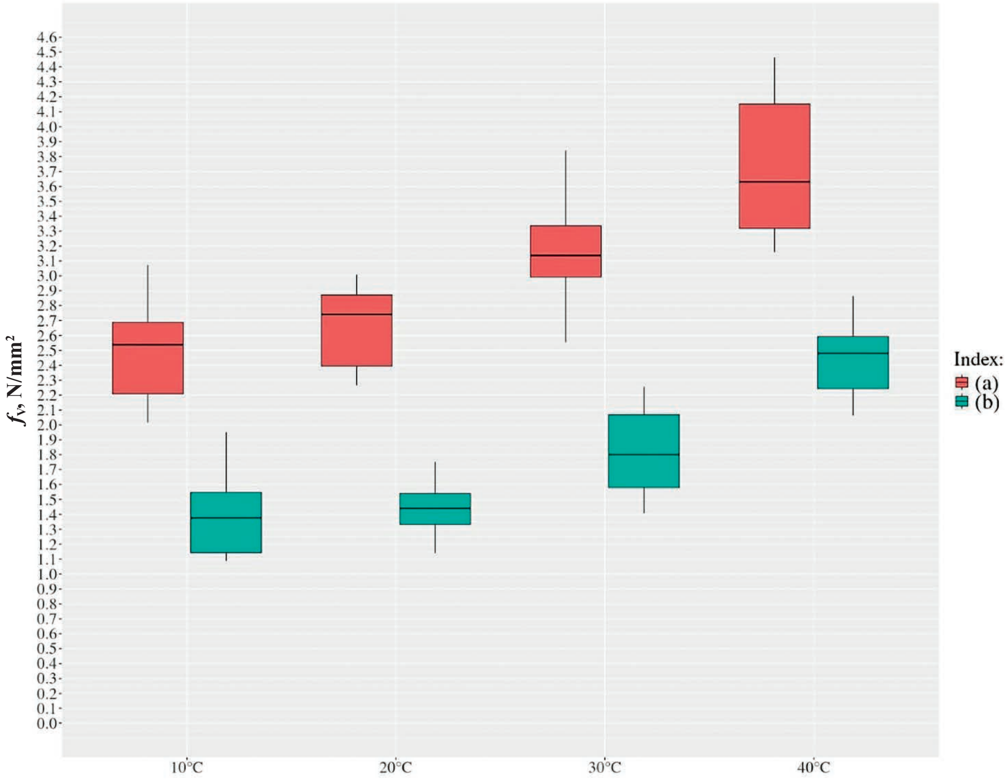

Figure 6 Influence of adhesive temperature on bonding strength in a dry state (a) and after immersion in boiling water for one hour (b)
Slika 6. Utjecaj temperature ljepila na čvrstoću lijepljenja furnirske ploče u suhom stanju (a) i nakon jednosatnog potapanja u kipućoj vodi (b)

alcohol-soluble PF resin can be assumed to be in the range of 30-40 °C.

4 CONCLUSIONS
4. ZAKLJUČAK

The use of alcohol-soluble PF resin for the production of plywood showed very high strength characteristics of the adhesive joint. The established results strongly indicate that these properties apply both to the dry state and to treatment in boiling water. This is of considerable interest when plywood is subjected to adverse weather conditions.

The strength of the adhesive joints rised almost linearly with increasing pressing time and temperature from 135 to 155 °C.

The established optimal mode factors after the conducted research were: temperature of 145 °C, adhesive spread of 150 g/m^2 and initial temperature of the glue 30-40 °C.

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