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Effects of Silane Treatment on Physical and Mechanical Properties of Particleboards Prepared with Urea Formaldehyde

Učinci obrade silanom na fizička i mehanička svojstva ploče iverice izrađene s urea-formaldehidnim ljepilom

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ABSTRACT • *In this study, the effect of silane on particleboard physical and mechanical properties was investigated. Silane was mixed in particleboards with two different methods. In the first method, silane was mixed with wood chips namely as a pretreatment of wood chips. In the second method, silane was mixed with urea-formaldehid adhesive. The amounts of silane used in both methods were 1 %, 2 % and 3 % of the particles based on the dry weight. Urea formaldehyde with 65 % solid content was used as adhesive. Ammonium sulfate (1 %) was added as a hardener to the used adhesive. The target density of three-layer particleboards manufactured was 550 kg/m^3. The properties of particleboards evaluated include thickness swelling, bending properties and internal bond strength. Results indicate that particleboards thickness swelling values increase with the use of silane. Overall, pretreatment of the wood particles provided better mechanical properties than the addition of silane to the adhesive solution. Flexure strength and flexure modulus of particleboards were significantly improved by use of silane. Pretreatment of particles with 2 % silane yielded the best improvement of modulus of rupture and modulus of elasticity. For the particleboards prepared with pretreated wood chips with silane, the highest increase of the flexure modulus and flexure strength was 20 % and 40 %, respectively. Pretreatment of wood particles with 3 % of silane nearly doubled internal bond strength of the particleboards. Silane pretreatment can be an alternative method for the improvement of particleboard mechanical properties.*

KEYWORDS: *silane; particleboard; physical and mechanical properties*

SAŽETAK • *U ovom je istraživanju ispitivan učinak silana na fizička i mehanička svojstva ploče iverice. Obrada ploče iverice silanom provedena je primjenom dviju metoda. U prvoj metodi silan je pomiješan s drvnim iverjem, što je ujedno bila predobrada drvnog iverja. U drugoj metodi silan je pomiješan s urea-formaldehidnim ljepilom. Količine silana upotrijebljenoga u obje metode bile su 1 %, 2 % i 3 % s obzirom na sadržaj suhe tvari. Kao ljepilo je rabljeno urea-formaldehidno ljepilo sa 65 % suhe tvari. Ljepilu je dodan amonijev sulfat (1 %) kao otvrdnjivač.*

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Ciljana gustoća proizvedene troslojne ploče iverice bila je 550 kg/m³. Istražena svojstva iverice obuhvatila su debljinsko bubrenje, savijanje i čvrstoću raslojavanja. Rezultati pokazuju da se pri upotrebi veće količine silana vrijednost debljinskog bubrenja ploče iverice povećava. Prethodna obrada drvnog iverja rezultirala je boljim mehaničkim svojstvima iverice nego dodavanje silana ljepilu. Čvrstoća na savijanje i modul savitljivosti ploče iverice znatno su poboljšani primjenom silana. Predobradom iverja s 2 % silana postignuto je najbolje poboljšanje modula loma i modula elastičnosti. Za ploču ivericu pripremljenu s drvnim iverjem prethodno obrađenim silanom najveće povećanje modula savitljivosti i čvrstoće na savijanje iznosilo je 20 %, odnosno 40 %. Predobrada drvnog iverja s 3 % silana gotovo je udvostručila čvrstoću raslojavanja iverice. Predobrada silanom može biti alternativna metoda za poboljšanje mehaničkih svojstava ploče iverice.

KLJUČNE RIJEČI: *silan; ploča iverica; fizička i mehanička svojstva*

1 INTRODUCTION
1. UVOD

Particleboards which are commonly used in furniture construction are one of the oldest engineered wood base materials. Physical and mechanical properties of particleboards are affected by a variety of factors such as particle geometry, wood species, density, adhesive type and ratio, etc. (Sanabria *et al.*, 2013; Istek and Ozlusoylu, 2017; 2019). Urea formaldehyde (UF) adhesive is the most commonly used adhesive in the particleboard industry due to some advantages such as cheapness, availability, easy curing, etc. Formaldehyde emission is the most important issue that the manufacturers and users encounter. Properties of urea formaldehyde (UF)-bonded particleboards are usually inferior (Han *et al.*, 1998) comparing to the particleboards manufactured using phenol (PF) or melamine formaldehyde (MUF) isocyanate (Istek *et al.*, 2020). The dimensional stability of particleboards can be improved by using water resistance adhesives such as isocyanate (Han *et al.*, 1998), water repellents, higher adhesive ratio (Ayrilmis *et al.*, 2012) or chemical modification (Clemons *et al.*, 1992; Rowell *et al.*, 1995; Mahlberg *et al.*, 2001). Some pretreatments are applied to the particles such as cold or hot water soaking (Zheng *et al.*, 2006; Pan *et al.*, 2007; Hartono *et al.*, 2018). NaOH (Fenghu *et al.*, 1993; Ishak *et al.*, 2013), enzyme (Zhang *et al.*, 2003) is also found to be effective in order to improve some physical and mechanical properties of particleboards manufactured using urea-formaldehyde. Physical and mechanical properties of the particleboards can also be altered by coating materials applied (Norvydas and Minelga, 2006). Liquified wood (Cuk *et al.*, 2011) and nano cellulose (Veigel *et al.*, 2012; Leng *et al.*, 2017) were also used as modifiers for adhesive and found promising for improvement of particleboard properties.

Silane is known to be an efficient coupling agent mostly used in adhesive formulations (Han, 1998; Ishak *et al.*, 2013; Istek *et al.*, 2016). Although silane treatment was found to be effective on some physical and mechanical properties of urea bonded reed and wheat straw particleboards (Han *et al.*, 1998), and phenol bonded rice husk particleboards (Sawawi *et al.*, 2018), negative effects were reported on urea bonded oriented strand lumber (Taghiyari *et al.*, 2017). Lower wettability, superior hardness and improved roughness of poplar–wheat straw particleboards were reported by using silane coupling agent (Hafezi and Doosthoseini, 2014). Another study revealed that using silane in fiberboard production caused color darkening and surface roughness (Istek *et al.*, 2016). According to Kloeser *et al.* (2007), silanes have lowering effect on formaldehyde emission from wood based panels.

In this study, silane was used in both pretreatment of particles and mixture of urea formaldehyde adhesive, and its effects on some physical and mechanical properties of the particleboards manufactured were investigated.

2 MATERIALS AND METHODS
2. MATERIJALI I METODE

Particles consisting of pine (85 %) and poplar (15 %) wood and urea formaldehyde adhesive were provided from a local particleboard factory (Masstaş Aş, Mudurnu, Bolu, Turkey). Urea formaldehyde (E2 class) with a solid content of 65 %, gel time of 35 s, viscosity of 450 cp, density of 1.281 g/cm³, pH of 8.2 was used. The amount of adhesive was 11 % and 12 % of the particles based on the dry weight. As a hardener, 1 % ammonium sulfate (solid content of 33 %) was mixed with the adhesive used. Silane was purchased from Wacker Company in the form of Geniosil® GF 9 that has a bridge building property.

Particleboards measuring 18 mm × 600 mm × 600 mm were prepared in the laboratory conditions. Two approaches were considered in the production of experimental particleboards. In the first, wood particles were pretreated with silane. Then, adhesive was sprayed to the particles until a homogeneous distribution was obtained. In the second, silane was mixed with the adhesive solution and then sprayed on the particles. After mixing, mats were placed in a steel frame by hand and pressed for 4 minutes at the temperature of 190 °C. The target density of the experimental panels was 550 kg/m³. After pressing, experimental boards were conditioned in the laboratory climate at approximately +20 °C, relative humidity of 65 %.

Table 1 Experimental design used in the study
Tablica 1. Dizajn eksperimenta u ovom istraživanju

Treatment *Obrada*	T1	T2	T3	T4	T5	T6	T7	T8	T9	T10	T11	T12	T13	T14
Treatment type *Vrsta obrade*	-	**Particles treated** *Obrada drvnog iverja*			**Mixed with adhesive** *Miješanje s ljepilom*			-	**Particles treated** *Obrada drvnog iverja*			**Mixed with adhesive** *Miješanje s ljepilom*		
Amount of silane used, % *količina silana, %*	-	1	2	3	1	2	3	-	1	2	3	1	2	3
Amount of adhesive used, % *količina ljepila, %*	11	11	11	11	11	11	11	12	12	12	12	12	12	12

After conditioning, samples were cut to the required size in order to determine some physical and mechanical properties. Experimental design used in the study is presented in Table 1.

The following physical and mechanical properties were determined using the applicable standards: apparent density (TSE EN 323, 1999); moisture content (*MC*) (TSE EN 322, 1999); thickness swelling (*TS*) (Figure 1) after 24 hour of water immersion (TSE EN 317, 1999); internal bond strength (*IBS*) (TSE EN 319, 1999), (Figure 2); modulus of elasticity (*MOE*) (TSE EN 310, 1999) and modulus of rupture (*MOR*) (TSE EN 310, 1999), (Figure 2) in bending.

Ten replicates were used for each test and the obtained data were subjected to an analysis of variance. Experimental results were analyzed using ANOVA tests to identify their statistical significance. Duncan's multiple range tests were performed in order to find the least significant difference between all the variables.

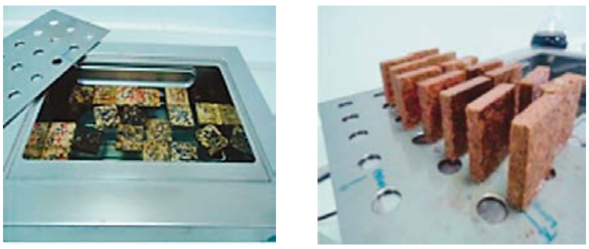


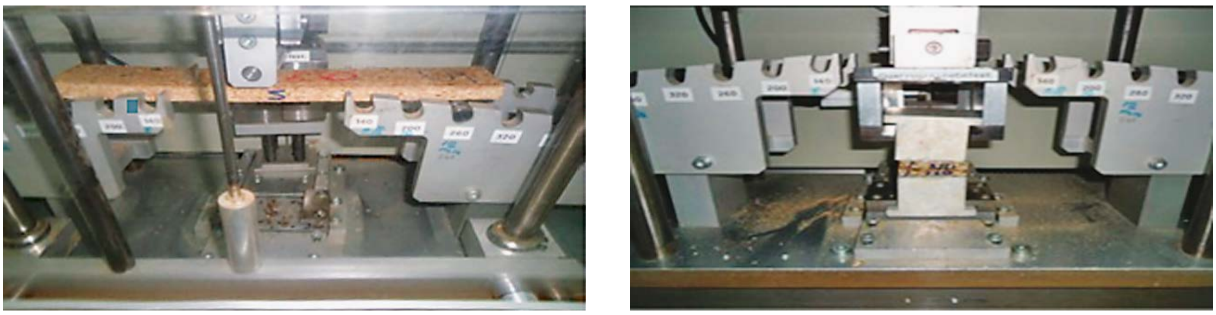
Figure 1 Thickness swelling
Slika 1. Debljinsko bubrenje

3 RESULTS AND DISCUSSION
3. REZULTATI I RASPRAVA

Table 2 shows some physical and mechanical properties of the laboratory manufactured particleboard samples. Density of the experimental particleboards ranges from 521 to 584 kg/m^3 and was not significantly different among the tested group. The use of silane did not significantly alter the density of the particleboards manufactured. Thickness swelling of the particleboards varies between 11.5 % and 30.9 % (Figure 3).

Test results indicate that both the amount of adhesive and silane used influence the thickness swelling of the particleboards. Particleboards (T8) without silane had the lowest thickness swelling values (11.7 %). After 24 hours of water immersion, thickness swelling values were the highest for the treatment T5 with 1 % pretreated particles and 11 % silane. It was considered that T5 has the lowest density (512 kg/m^3) among the other samples. The amount of adhesive has greater effect on thickness swelling values than silane used. The addition of a hydrophobic substance such as waxes usually mixed with the adhesive solution in the production of particleboards would change water affiliation, and thus lower thickness swelling values.

Increasing amount of adhesive lowers thickness swelling values of particleboards as expected (Ayrilmis and Nemli, 2017). The use of silane seems to have more apparent effect on particleboards manufactured



a) b)

Figure 2 a) Bending strength, b) Internal bond strength
Slika 2. a) Čvrstoća na savijanje, b) čvrstoća raslojavanja

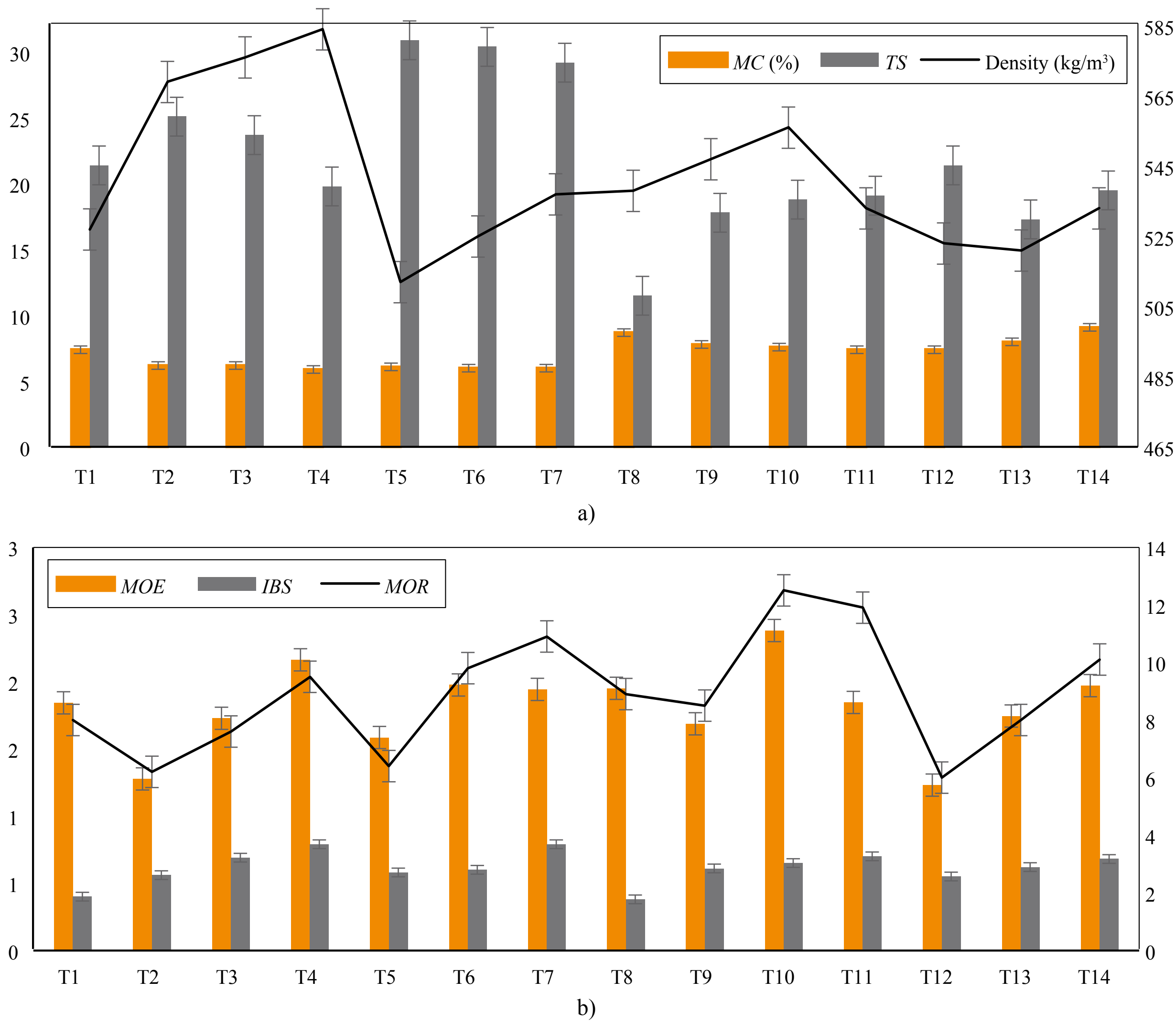


Figure 3 a) Physical and b) mechanical properties of particleboards
Slika 3. a) Fizička svojstva ploče iverice, b) mehanička svojstva ploče iverice

using 11 % adhesive than those manufactured using 12 % adhesive. Higher thickness swelling values may be attributed to the buffering effect of silane. The effect of silane on thickness swelling values of wood based panels in the literature is controversial. After the use of silane, increase of TS was reported for OSL manufactured using poplar and urea formaldehyde (Taghiyari *et al.*, 2017) and particleboards manufactured using 0.5 mesh Kelampayan particles (Ishak *et al.*, 2013). An investigation on reed and wheat straw particleboards (Han *et al.*, 1998) presented that the use of silane coupling agents improves TS values. A study conducted by Kharazipour *et al.* (2009) shows that the use of silane in the manufacture of MDF reduces the 2-hour TS values by half. Improvement in TS for rice husk particleboards was also reported when silane was used with phenol formaldehyde (Sawawi *et al.*, 2018).

Modulus of elasticity in bending values of tested particleboards varies between 1230 N/mm^2 and 2380 N/mm^2. Bending strength of the panels ranges from 6 N/mm^2 to 12.5 N/mm^2. ANOVA results indicate that bending properties among the treatments differ, but all results are in the same group (A and AB). The highest values of bending properties were obtained by treatment T10 in which particles were pretreated with 2 % silane and 12 % adhesive used (Figure 3). It seems that pretreatment of particles yields better bending *MOE* and *MOR* values than when silane is mixed with the adhesive solution. 1 % increase in the amount of adhesive causes approximately 5 % increase in *MOE,* while the increase in *MOR* is more than 10 %. Comparing with the control groups (T1 and T8), the increase in bending properties with the addition of silane is more efficient (more than 20 % for stiffness, 40 % for bending strength) than adhesive quantity increases (from11 % to 12 %). Higher bending properties are expected with the use of a higher amount of adhesive (Rangavar *et al.*, 2016; Ayrilmis and Nemli, 2017) but interaction of silane with the adhesive used induces excelsior bending properties.

The static bending strength requirement for general-purpose particleboards (TS EN 312, 2005) is 11.5 N N/mm^2. Treatment T10, manufactured with pretreated silane, exceedingly meets the minimum values of bending properties required by the standards.

Table 2 Some physical and mechanical properties of particleboard samples
Tablica 2. Fizička i mehanička svojstva uzoraka ploče iverice

Treatment *Obrada*	**Density, kg/m³** *Gustoća,* kg/m³	***MC*, %**	***TS*, %**	***MOE*, N/mm²**	***MOR*, N/mm²**	***IBS*, N/mm²**
T1	527	7.4	21.4 AB*	1842 AB	8 AB	0.4 A
T2	569	6.2	25.1 AB	1276 A	6.2 A	0.56 A
T3	576	6.2	23.7 AB	1727 A	7.6 A	0.69 AB
T4	584	5.9	19.8 AB	2162 AB	9.5 AB	0.74 AB
T5	512	6.1	30.9 B	1584 A	6.4 A	0.58 A
T6	525	6	30.4 AB	1975 AB	9.8 AB	0.6 AB
T7	537	6	29.2 AB	1942 AB	10.9 AB	0.79 B
T8	538	8.7	11.5 A	1950 AB	8.9 AB	0.38 A
T9	547	7.8	17.8 A	1586 A	8.5 AB	0.61AB
T10	556	7.6	18.8 A	2380 B	12.5 B	0.65 AB
T11	533	7.4	19.1 A	1845 AB	11.9 AB	0.7 AB
T12	523	7.4	21.4 AB	1230 A	6 A	0.55 A
T13	521	8	17.3 A	1744 AB	8 A	0.62 AB
T14	533	9.1	19.5 AB	1970 AB	10.1 AB	0.68 AB

*Indicates Duncan grouping / *označava Duncanovo grupiranje*

Internal bond strength of control groups is not significantly different. Internal bond strength is increased by the amount of adhesive used and addition of silane. The minimum requirement of internal bond strength for general-purpose particleboards (TS EN 312, 2005) is 0.24 N/mm². All of the boards manufactured in the study exceed the minimum requirements.

Internal bond strength is nearly doubled by the treatment groups of T7 and T11, manufactured with pretreated particles with 3 % silane. Similar results were also reported for internal bond strength of MDF (Kloeser *et al.*, 2007; Kloeser, 2010), particleboards manufactured using Kelampayan particles (Ishak *et al.*, 2013).

4 CONCLUSIONS
4. ZAKLJUČAK

This study investigated the influence of silane pretreatment of particles and silane addition to the adhesive solution on some physical and mechanical properties of particleboards. Contrary to previous studies, silane negatively affected the thickness swelling and moisture content of particleboards in this study. It is considered that particleboard density played an important role in this difference. In this study, lower particleboard density was observed than in controversial studies. Mechanical properties of the particleboards improved with silane because silane increased the internal bond across the particle and adhesive in the board. *MOE*, *MOR* and *IBS* test results show that silane increases the internal bonding of particleboard because it joins the particles to each other very effectively. Pretreatment of particles was found to be a more efficient approach than silane addition to the adhesive solution. Water repellents and manufacturing higher density board could yield better board properties. Silane could be used for improvement of particleboard properties, and it could be an alternative to other pretreatments or chemical additives.

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