

Engineering the Properties of Eco-Friendly Medium Density Fibreboards Bonded with Lignosulfonate Adhesive

Dizajniranje svojstava ekološki prihvatljivih ploča vlaknatica srednje gustoće proizvedenih uporabom lignosulfonatnog ljepila

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ABSTRACT • Free formaldehyde emissions from wood-based panels, especially in indoor applications, pose serious risks to human health at certain concentrations. Prolonged exposure to formaldehyde can cause adverse health effects including eye, nose and throat irritation, other respiratory symptoms and cancer. As a consequence, new formaldehyde emission limits for composite wood products were established in Europe, USA and Japan. This, together with the stricter environmental legislation are the main driving factors for shifting the scientific and industrial interest from the traditional formaldehyde-based synthetic resins to the new bio-based adhesives for production of eco-friendly wood-based panels. The lignin-based products are one of the most prospective ecological alternatives to the traditional formaldehyde resins. The main interest in lignin is due to its phenolic structure with several favourable properties for the formulation of wood adhesives such as high hydrophobicity and low polydispersity.

The present article is aimed at studying the possibilities for using lignosulfonate as an adhesive for the production of eco-friendly MDF. Regression models describing the impact of lignosulfonate concentration and hot pressing temperature on the exploitation properties of MDF panels were developed. The individual and combined impact of both factors was analysed in order to determine the optimal exploitation properties of the panels.

Keywords: eco-friendly MDF; lignosulfonate; physical and mechanical properties; bio-based adhesives; wood-based panels

SAŽETAK • Emisija slobodnog formaldehida iz ploča na bazi drva, posebice primijenjenih u unutarnjim prostorima, u određenim je koncentracijama ozbiljan rizik za zdravlje ljudi. Dulja izloženost formaldehydu može uzrokovati znatne zdravstvene probleme, uključujući iritaciju očiju, nosa i grla, druge respiratorne simptome i rak. Stoga su u Europi, SAD-u i Japanu određene nove granice emisije formaldehida za kompozitne proizvode od drva. To je, zajedno sa strožim zakonodavstvom o okolišu, bio glavni poticaj za prebacivanje znanstvenoga i industrijskog fokusa s tradicionalnih sintetičkih smola temeljenih na formaldehydu na nova prirodna ljepila za proizvodnju ekološki prihvatljivih ploča na bazi drva. Proizvodi na bazi lignina jedna su od najperspektivnijih ekoloških alter-

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nativa tradicionalnim formaldehidnim smolama. Glavni razlog zanimanja za lignin jest njegova fenolna struktura s nekoliko povoljnih svojstava za formulaciju ljepila za drvo poput visoke hidrofobnosti i niske polidisperznosti. Cilj ovog rada jest proučavanje mogućnosti uporabe lignosulfonata kao ljepila za proizvodnju ekološki prihvatljivih ploča vlaknatica srednje gustoće (MDF ploča). Razvijeni su regresijski modeli koji opisuju utjecaj koncentracije lignosulfonata i temperature prešanja na svojstva MDF ploča. Analiziran je pojedinačni i kombinirani utjecaj obaju elemenata kako bi se odredila optimalna svojstva ploča.

Ključne riječi: ekološki prihvatljive MDF ploče; lignosulfonat; fizička i mehanička svojstva; prirodna ljepila; ploče na bazi drva

1 INTRODUCTION

1. UVOD

The production of medium density fibreboards (MDF) is one of wood-based industries with the fastest growth rate. Production in the European Panel Federation member countries grew by 2 % in 2016 to 12 million m³ and in the broader European subregion it grew by 2.6 %, to 17.5 million m³ (FAO 2017, European Panel Federation 2017). One of the main technical barriers and disadvantages associated with this production is the free formaldehyde emission that poses serious risks to human health at certain concentrations (Carvalho *et al.*, 2012; Mantanis *et al.*, 2018). This problem has been extensively studied in the recent years (Athanasosiadou *et al.*, 2009; Pizzi, 2016) and one of the most advanced solutions is the use of formaldehyde-free bio-based adhesives, which do not significantly increase the production costs of wood-based panels and at the same time maintain their exploitation properties (Nordström *et al.*, 2017, Pizzi, 2003; Pizzi, 2006; El Mansouri *et al.*, 2007; Sepahvand *et al.*, 2018; Hemmilä *et al.*, 2017).

The lignin-based products, including lignosulfonates, are one of the most prospective ecological alternatives to the traditional formaldehyde adhesive systems, used in the wood panel industry (Shimatani *et al.*, 1994; El Mansouri *et al.*, 2007; Nordström *et al.*, 2017; Pizzi, 2016; Yotov *et al.*, 2017). Lignosulfonates have good bonding properties when applied to wood fibres; the main barrier for their wide industrial application is the worsened water resistance of fabricated panels (Dimitrescu *et al.*, 2009). This issue can be partially resolved by adding lignosulfonates to other adhesive systems (Nasir *et al.*, 2014; Savov *et al.*, 2017; Hemmilä *et al.*, 2019) or by modifying the parameters for the production of panels (Antov *et al.*, 2019; Savov *et al.*, 2019).

Analytical study on engineering the properties of eco-friendly MDF panels produced in laboratory conditions using lignosulfonates as adhesives is presented in this article. Two main factors were studied – the concentration of adding lignosulfonate in the wood-fibre mass and hot pressing temperature.

2 MATERIALS AND METHODS

2. MATERIJALI I METODE

In order to design the properties of eco-friendly MDF, panels were produced at three different lignosulfonate concentrations – 20 %, 30 % and 40 %, and three hot pressing temperatures – 200 °C, 210 °C and

220 °C. The panels were produced with a thickness of 6 mm and a density of 850 kg/m³. The experimental plan is presented in Table 1.

Table 1 Experimental plan
Tablica 1. Plan istraživanja

No.	MDF panel density ρ , kg/m ³ <i>Gustoća MDF ploča ρ, kg/m³</i>	Lignosulfonate concentration K , % <i>Koncentracija lignosulfata K, %</i>	Hot pressing temperature T , °C <i>Temperatura prešanja T, °C</i>
1	850	20	200
2	850	20	210
3	850	20	220
4	850	30	200
5	850	30	210
6	850	30	220
7	850	40	200
8	850	40	210
9	850	40	220

Factory-produced wood-fibre mass with the following composition – common beech (*Fagus sylvatica* L.) – 57 %, European oak (*Quercus robur* L.) – 35 %, and white poplar (*Populus alba* L.) – 8 %, was used for the purpose of the study. The mass was dried to 11 % water content. The pulp freeness, determined by the Schopper-Riegler method, was 11° ShR. The bulk density of the mass was 29 kg/m³. The calcium lignosulfonate content was 15 % and had the following characteristics: calcium - up to 6 %; reduced sugars – 7 %; ash content – 14 %; dry content – 93 %; acidic factor in 10 % solution - pH = 4.3 ± 0.8; bulk density - 550 kg/m³.

Wood fibres were mixed with the lignosulfonate and paraffin for 50 s in a high-speed glue blender (850 min⁻¹). The hot pressing was performed on a laboratory press type PMC ST 100, Italy. The applied pressing regime of MDF was as follows: in the first stage, the pressure was increased for 20 s to 3.0 MPa and maintained for 20 % of the whole pressing cycle, then the pressure was evenly decreased for 10 s to 1.2 MPa and maintained for a duration of 30 % of the whole pressing cycle; the third (last) pressing period was carried out at the pressure of 0.6 MPa for 50 % of the whole cycle. The pressing factor was 90 s/mm¹. The physical and mechanical properties of the obtained MDF panels were determined in accordance with the requirements of the respective EN standards (EN 310; EN 317; EN 319; EN 322 and EN 323). The mechanical properties of MDF panels were determined on a universal testing machine Zwick/Roell Z010.

On this basis the following regression equation in a coded form was derived:

$$\hat{Y} = B_0 + B_1 \cdot X_1 + B_2 \cdot X_2 + B_{12} \cdot X_1 \cdot X_2 + B_{11} \cdot X_1^2 + B_{22} \cdot X_2^2 \quad (1)$$

where \hat{Y} is the predicted value of the respective MDF output parameter, X_1 is the lignosulfonate concentration in a coded form and X_2 is the hot pressing temperature in a coded form.

A standard methodology was used to determine the adequacy of the model by calculating the Fisher criterion and comparing it with the table values. Variance relation $F_{calc} = \frac{S_y^2}{S_\varepsilon^2}$ was calculated, where S_y^2 is the residual variance and S_ε^2 is the variance of random disturbance determined on the basis of the variance under the null experiment (at 30 % lignosulfonate concentration and 210 °C hot pressing temperature). The testing was carried out on 8 test samples for each parameter and variational and statistical processing of the results was made to test the adequacy of the model, encoded by the following formula:

$$\tilde{X}_i = X_i \cdot \Delta_i + X_{i0} \quad (2)$$

where \tilde{X}_i is the natural meaning of the factor, Δ_i is the variation interval and X_{i0} is the variation interval median.

The accuracy of the applied model was determined by the multiple correlation coefficient.

3 RESULTS AND DISCUSSION

3. REZULTATI I RASPRAVA

The summarised results of the exploitation properties of the laboratory produced MDF panels at different lignosulfonate concentrations and hot pressing temperature are presented in Table 2.

Data about the tests in the null experiment are presented in Table 3.

All derived equations correspond to the critical value $F(0.005; 3; 7) = 4.35$, and the values for the respective models are: for the water absorption – 2.37; for the swelling in thickness – 3.17; for the bending strength – 3.85; for the modulus of elasticity in bending – 3.02; and for the internal bond strength – 4.21.

The coded form of the regression equation for the water absorption of MDF panels is:

$$\hat{A} = 57.20 - 5.02 \cdot X_1 - 1.63 \cdot X_2 + 2.35 \cdot X_1^2 + 0.2 \cdot X_2^2, \% \quad (3)$$

where X_1 is the lignosulfonate concentration and X_2 is the hot pressing temperature in coded form. The equation is characterised by the coefficient of determination $R^2 = 0.96$.

From Eq. 3, it can be concluded that the effect of lignosulfonate concentration was significantly stronger in the studied range. The impact of the factor at the variation range from 20 to 40 % was about 2.6 greater than the effect of hot pressing temperature within the

Table 2 Results of physical and mechanical properties of eco-friendly MDF panels

Tablica 2. Rezultati ispitivanja fizičkih i mehaničkih svojstava ekološki prihvatljivih MDF ploča

Panel No. <i>Broj ploče</i>	Density ρ , kg/m ³ <i>Gustoća ρ, kg/m³</i>	Water absorption A , % <i>Upijanje vode A, %</i>	Swelling in thickness G_t , % <i>Bubrenje G_t, %</i>	Bending strength f_m , N/mm ² <i>Čvrstoća na savijanje f_m, N/mm²</i>	Modulus of elasticity in bending E_m , N/mm ² <i>Modul elastičnosti pri savijanju E_m, N/mm²</i>	Internal bond strength f_v , N/mm ² <i>Čvrstoća na raslojavanje f_v, N/mm²</i>
1.	855±7	74.8±2.7	24.1±1.0	22.75±0.4	2894±97	0.62±0.005
2.	841±9	64.8±1.9	23.0±0.6	20.89±0.7	2655±130	0.66±0.025
3.	831±8	62.5±1.8	22.0±0.7	24.79±0.6	3308±130	0.68±0.026
4.	849±5	58.3±1.7	21.8±0.8	32.08±0.4	3734±115	0.70±0.009
5.	847±7	57.8±2.3	19.8±0.6	30.54±1.0	3853±188	0.71±0.026
6.	856±8	55.5±2.4	19.5±0.5	30.58±1.1	3956±129	0.71±0.029
7.	842±8	56.5±2.7	20.8±1.0	30.33±1.3	3676±85	0.74±0.015
8.	854±5	53.7±2.5	19.6±0.4	32.08±0.8	3440±92	0.72±0.011
9.	854±5	53.4±1.9	18.5±0.8	26.61±0.8	3613±96	0.64±0.027

Table 3 Initial panel property values

Tablica 3. Početne vrijednosti svojstava ploče

Properties <i>Svojstva</i>	Sample No. <i>Broj uzorka</i>							
	1	2	3	4	5	6	7	8
Water absorption A , % / upijanje vode A , %	55.67	47.48	60.45	57.22	53.11	43.51	55.24	54.78
Swelling in thickness G_t , % / bubrenje G_t , %	19.93	14.77	16.48	17.07	20.69	18.28	20.58	20.38
Bending strength f_m , N/mm ² <i>čvrstoća na savijanje f_m, N/mm²</i>	24.5	28.2	26.1	26.8	28.2	22.1	28.4	28.60
Modulus of elasticity E_m , N/mm ² <i>modul elastičnosti pri savijanju E_m, N/mm²</i>	3650	3550	3390	3680	3560	3150	3940	3980
Internal bond strength f_v , N/mm ² <i>čvrstoća na raslojavanje f_v, N/mm²</i>	0.828	0.679	0.615	0.674	0.677	0.699	0.811	0.737

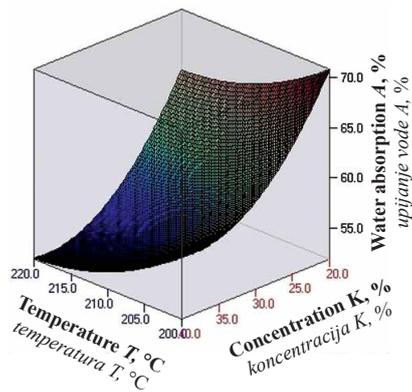


Figure 1 Variation of water absorption of eco-friendly MDF panels depending on lignosulfonate concentration and hot pressing temperature

Slika 1. Varijacije upijanja vode ekološki prihvatljivih MDF ploča ovisno o koncentraciji lignosulfonata i temperaturi prešanja

range from 200 to 220 °C. This can be attributed to the selected relatively high hot pressing temperatures above 200 °C. Therefore, water absorption can be better designed by varying the concentration of adding lignosulfonate in the wood-fibre mass. The graphic representation of this dependence after encoding the equation is presented in Figure 1.

It can be concluded that an improvement in water resistance (decreased water absorption) was determined when lignosulfonate concentration was increased from 20 % to 40 % and hot pressing temperature was increased from 200 °C to 220 °C, respectively. The water absorption values were significantly improved when the lignosulfonate concentration was increased from 20 % to 35 % and the hot pressing temperature – from 200 °C to 210 °C.

The coded form of the regression equation for the swelling in thickness of the laboratory produced MDF panels is:

$$\hat{G}_t = 20.10 - 1.79 \cdot X_1 - 1.21 \cdot X_2 + 0.09 X_1 \cdot X_2 + 1.05 \cdot X_1^2 + 0.41 \cdot X_2^2, \% \quad (4)$$

where X_1 is the lignosulfonate concentration and X_2 is the hot pressing temperature in coded form. The equation is characterised by a coefficient of determination $R^2 = 0.95$.

Decreased swelling in thickness of MDF panels was determined with increasing the concentration of lignosulfonate solution and hot pressing temperature. Again, lignosulfonate concentration was the more significant factor in the studied variation range. However, the effects of both studied factors were comparable. Strong quadratic dependence was determined between the swelling in thickness of MDF panels and lignosulfonate concentration.

The variation of swelling in thickness of MDF panels depending on lignosulfonate concentration and hot pressing temperature is presented in Figure 2.

The analysis of the obtained results of this property is similar to the water absorption of the panels. Significantly greater relative improvement was determined with increasing the lignosulfonate concentration

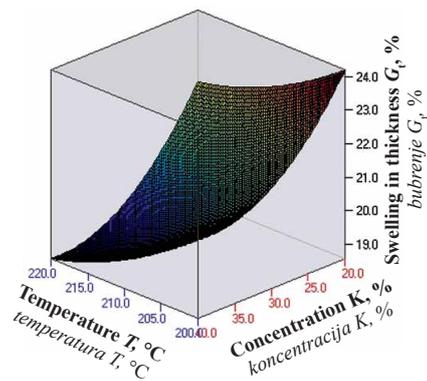


Figure 2 Variation of swelling in thickness of eco-friendly MDF panels depending on lignosulfonate concentration and hot pressing temperature

Slika 2. Varijacije bubrenja ekološki prihvatljivih MDF ploča ovisno o koncentraciji lignosulfonata i temperaturi prešanja

up to 35 % and less with increasing the concentration from 35 % to 40 %. Regarding the hot pressing temperature, the relatively greater improvement was determined with increasing the temperature up to 210 °C.

The coded form of the regression equation for the bending strength is as follows:

$$\hat{f}_m = 31.54 + 4.15 \cdot X_1 + 0.2 \cdot X_2 - 2.52 X_1 \cdot X_2 - 5.54 \cdot X_1^2 - 0.07 \cdot X_2^2, \text{ N} \cdot \text{mm}^2 \quad (4)$$

where X_1 is the lignosulfonate concentration and X_2 is the hot pressing temperature in coded form. The coefficient of determination of Eq. 4 is $R^2 = 0.90$.

The effect of lignosulfonate concentration on this property was significantly greater than the effect of hot pressing temperature. The quadratic dependence between the lignosulfonate concentration and bending strength of the panels was strongly pronounced. A strong impact of the interaction of both studied factors on bending strength values was determined in comparison with the water absorption and swelling in thickness of the laboratory produced MDF panels. In other words, processes of destruction of wood components occur at the relatively higher hot pressing temperature and lignosulfonate concentration and low moisture content of the pressed material, which results in increased panel brittleness.

The graphical representation of the dependence of bending strength on lignosulfonate concentration and hot pressing temperature after encoding Eq. 4 is given in Figure 3.

Improvement of bending strength values was determined with increasing the lignosulfonate concentration from 20 % to 30 % and increasing the hot pressing temperature from 200 °C to 220 °C. Further increase of hot pressing temperature and lignosulfonate concentration above 30 % resulted in deterioration of the studied property. This deterioration was even more strongly pronounced at concentrations above 35 %.

To achieve the EN 622-5 standard requirement of 27 N/mm² for MDF panels with general application in humid environment, the lignosulfonate concentration should be at least 25 %.

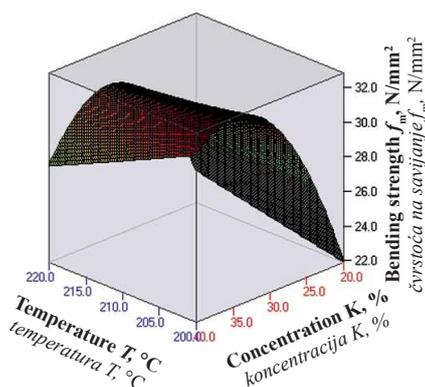


Figure 3 Variation of bending strength of eco-friendly MDF panels depending on lignosulfonate concentration and hot pressing temperature

Slika 3. Varijacije čvrstoće na savijanje ekološki prihvatljivih MDF ploča ovisno o koncentraciji lignosulfonata i temperaturi prešanja

The regression equation regarding the modulus of elasticity of MDF panels is as follows:

$$\hat{E}_m = 3753 + 385 \cdot X_1 + 168 \cdot X_2 - 228 \cdot X_1 \cdot X_2 - 656 \cdot X_1^2 + 141 \cdot X_2^2, \text{ N} \cdot \text{mm}^{-2} \quad (5)$$

where X_1 is the lignosulfonate concentration and X_2 is the hot pressing temperature in coded form. The coefficient of determination of equation (5) is $R^2 = 0.97$.

Similar to the bending strength, the modulus of elasticity in bending was more dependent on lignosulfonate concentration compared to the hot pressing temperature, i.e. the concentration factor is more appropriate for engineering the respective MDF property. The interaction of the studied factors resulted in decreased values of the modulus of elasticity in bending. The graphic representation of the dependence of the modulus of elasticity in bending on the lignosulfonate concentration and hot pressing temperature, after encoding Eq. 5, is presented in Figure 4.

Improvement of the modulus of elasticity in bending was determined with increasing the hot pressing temperature and lignosulfonate concentration up to 35 %. Further increase of temperature and concentration resulted in decreased values of the studied prop-

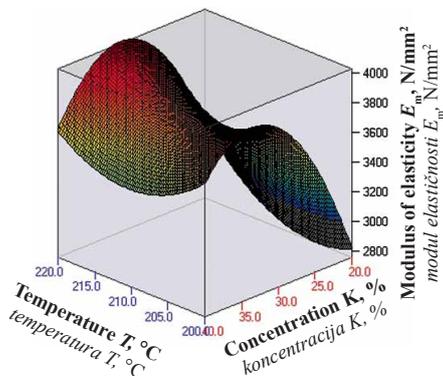


Figure 4 Variation of modulus of elasticity in bending of eco-friendly MDF panels depending on lignosulfonate concentration and hot pressing temperature

Slika 4. Varijacije modula elastičnosti pri savijanju ekološki prihvatljivih MDF ploča ovisno o koncentraciji lignosulfonata i temperaturi prešanja

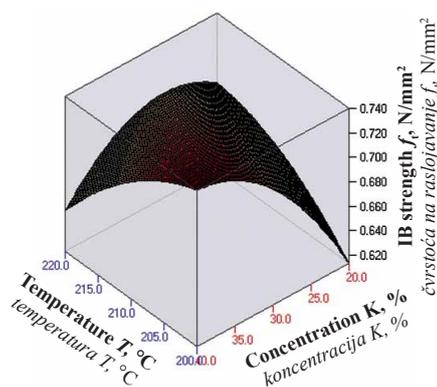


Figure 5 Variation of internal bond strength of eco-friendly MDF panels depending on lignosulfonate concentration and hot pressing temperature

Slika 5. Varijacije čvrstoće na raslojavanje ekološki prihvatljivih MDF ploča ovisno o koncentraciji lignosulfonata i temperaturi prešanja

erty. This can also be explained by the occurring destruction of some wood components under the impact of the increased lignosulfonate concentration, hot pressing temperature and pressing duration.

The regression equation of the internal bond strength of the panels is as follows:

$$\hat{f}_t = 0.705 + 0.033 \cdot X_1 + 0.005 \cdot X_2 - 0.014 \cdot X_1 \cdot X_2 - 0.013 \cdot X_1^2 + 0.002 \cdot X_2^2, \text{ N} \cdot \text{mm}^{-2} \quad (6)$$

where X_1 is the lignosulfonate concentration and X_2 is the hot pressing temperature in coded form. The coefficient of determination of Eq. 5 is $R^2 = 0.98$.

Separately, the studied factors had positive impact on the internal bond strength of MDF panels. Once again, the negative effect of their interaction was reported. The graphic representation of the dependence of the internal bond strength on the lignosulfonate concentration and hot pressing temperature, after encoding Eq. 6, is presented in Figure 5.

Improvement of internal bond strength values was determined at lignosulfonate concentration of up to 35 % and increase of the hot pressing temperature up to 220 °C. Further increase of hot pressing temperature resulted in decreased values of the studied property.

4 CONCLUSIONS

4. ZAKLJUČAK

The use of lignosulfonate as an adhesive is a perspective approach for producing eco-friendly MDF panels without harmful free-formaldehyde emissions. The exploitation properties of MDF panels can be successfully engineered by varying the lignosulfonate concentration from 20 % to 40 % and hot pressing temperature from 200 °C to 220 °C. The concentration of adding lignosulfonate solution to wood-fibre mass has a stronger impact on the panel properties within the determined variation range in comparison with the hot pressing temperature. This can be explained by the relatively high (more than 200 °C) hot pressing temperature and increased press time. For this reason, a deterioration of the strength properties of the panels is

observed with increasing the hot pressing temperature at lignosulfonate concentrations above 35 %. Decreasing the concentration below 30 % is not justified since this does not lead to additional utilisation of the lignosulfonate hydroxyl groups but requires increasing the hot pressing temperatures.

Despite the significant positive results achieved in the production of lignosulfonate-based MDF panels, there are some considerable drawbacks that should be noted, e.g. the difficulties in activating lignosulfonates and their binding to wood fibres, as well as the required substantial increase of pressing temperature and extended pressing time. The latter outlines the future trends of extensive research in the field of MDF production on the basis of lignosulfonate adhesives.

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