

Supplementation of Natural Tannins as an Alternative to Formaldehyde in Urea and Melamine Formaldehyde Resins used in MDF Production

Dodatak prirodnih tanina kao alternativa formaldehidu u urea-formaldehidnim i melamin-formaldehidnim ljepilima u proizvodnji MDF ploča

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ABSTRACT • The aim of this study was to evaluate some mechanical and physical properties of medium density fiberboards (MDF) made from different adhesive types. Five panel types were made from mixtures of tannin (T) to urea formaldehyde (UF) and tannin to melamine formaldehyde (MF), TUF_{10%}, TUF_{20%}, MF, TMF_{10%} and TMF_{20%} respectively. Besides, the effect of press temperatures (170 and 180 °C) on the curing of the formulations was studied. Some mechanical properties (modulus of rupture, modulus of elasticity and internal bond strength), physical properties (thickness swelling and water absorption) and formaldehyde emission of the resulting panels were determined. The incorporation of tannin decreases the formaldehyde emission of the panels. However, flexural properties and internal bond strength decreased with increasing tannin ratio. The results indicated that panels can be manufactured using tannin up to 10 % in UF resin without falling below the minimum EN Standard requirements of mechanical properties for general purpose MDF panels.

Key words: Tannin, MF, UF, Formaldehyde emission, MDF

SAŽETAK • Cilj rada bio je procijeniti određena mehanička i fizikalna svojstva ploča vlaknatica srednje gustoće (MDF ploča) proizvedenih primjenom različitih vrsta ljepila. Pet vrsta ploča proizvedeno je uz primjenu smjese tanina (T) i urea-formaldehidne smole (UF) te tanina i melamin-formaldehidne smole (MF) u različitim omjerima, označenih kao TUF10%, TUF20%, MF, TMF10% i TMF20%. Osim toga, proučavan je utjecaj temperature prešanja (170 i 180 °C) na otvrdnjavanje tako dobivenih smjesa ljepila. Upoznata su i neka mehanička svojstva

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proizvedenih MDF ploča (modul loma, modul elastičnosti i čvrstoća raslojavanja), njihova fizikalna svojstva (bubrenje u debljinu i upijanje vode) te emisija formaldehida. Uz dodatak tanina proizvedene ploče imaju manju emisiju formaldehida. Međutim, čvrstoća na savijanje i čvrstoća raslojavanja tih ploča smanjuju se s povećanjem udjela tanina u ljepilu. Rezultati su pokazali da ploče mogu biti proizvedene s ljepilima u kojima je urea-formaldehidnim smolama dodano do 10 % tanina a da ploče i dalje udovoljavaju minimalnim EN standardnim zahtjevima mehaničkih svojstava za MDF ploče opće namjene.

Ključne riječi: tanin, MF, UF, emisija formaldehida, MDF

1 INTRODUCTION

1. UVOD

Over the past few decades, as a result of environmental awareness, the problem of air pollution in residential buildings increased more and more concerns on indoor air pollutants such as formaldehyde and acetaldehyde to be released from building materials and products. The release of formaldehyde from the panels used for internal applications is one of the main factors that cause sick building syndrome known indoor. Thus, the problem of formaldehyde emissions is one of the most important aspects of research of urea formaldehyde resin that should be seriously and thoroughly investigated and studied. On the other hand, the demand for wood composites such as medium density fiberboard, oriented strand board (OSB), plywood, hardboard and veneer products has recently increased significantly all over the world (Pirayesh and Saadatnia, 2015). Solving old and new challenges and movement towards green chemistry will require human unity and creativity. A new approach will also be needed to make better and greater use of renewable resources, and increase the resilience and diversity of production systems (Salari *et al.*, 2013). A selection of bio wastes including wheat straw, sugarcane, sunflower seed hull, bamboo and palm have been successfully used in panel manufacturing and are already in the market under different trade names (Buyuksari *et al.*, 2010).

Urea formaldehyde resin is the most important type of adhesive for manufacturing wood based composites (Kim and Kim, 2005; Nemli *et al.*, 2008). Low price, good technological properties, absence of colors in cured polymer and easiness of application for a variety of curing conditions are the advantages of urea formaldehyde, while, formaldehyde emission and poor water resistance are its main disadvantages (Abdolzahdeh *et al.*, 2011; Papadopoulos and Hague, 2003). With rising economic standards, concerns about human health and the environment have been raised due to the increasing demand for wood based panels (Habibi, 2008). Formaldehyde (HCOH) is a suspected human carcinogen that is known to be released from pressed-wood products used in home construction, including products made with urea-formaldehyde (UF) resins (Khanjanzadeh *et al.*, 2013). Formaldehyde has also been found to produce nasal carcinomas in mice and rats after exposure to 14.1 and 5.6 ppm of formaldehyde, respectively, over a long period of time (Khanjanzadeh *et al.*, 2013). Formaldehyde emission from panels in service is caused by residual formaldehyde present in the UF bonded panels trapped as gas in the

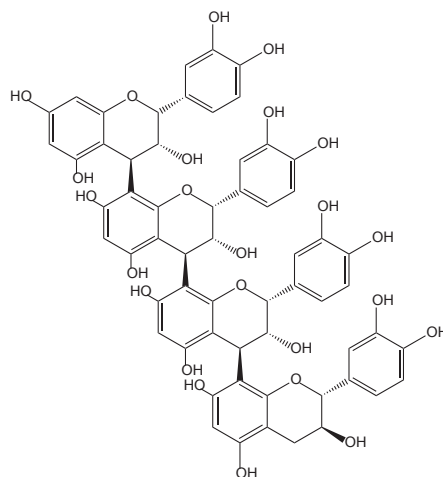


Figure 1 Condensed tannin
Slika 1. Kondenzirani tanin

structure and formaldehyde dissolved in the water present in the boards (Aydin *et al.*, 2006; Doosthosseini, 2002). UF is water based wood adhesive whose shortcomings are low efficiency, poor water resistance and formaldehyde emission (Doosthosseini, 2002). Due to similar reasons, other water-based adhesives, including protein and starch based adhesives, are also not effective to bond directly grass-based particleboards (Roumeli *et al.*, 2010)

Recently, there has been an interest in using tannin-based resins due to the more widespread availability of tannins, and the lower cost of tannin-based resins compared to CNSL. Tannins (Fig. 1) are naturally occurring phenolic compounds, which have been the subject of extensive research leading to the development of a wide range of industrial applications. The term “tannin” comes from the ancient Celtic word for oak, which still remains a popular source for tannins used in converting animal skin into leather (tanning). The objective of this study was to investigate the suitability of using natural tannins as an alternative to formaldehyde in urea and melamine formaldehyde resins used in MDF construction and to test the mechanical and physical properties of panels to determine if they have the required properties for general use.

2 MATERIAL AND METHODS

2. MATERIJAL I METODE

The raw material of this study included industrial wood fibers provided by a commercial fiberboard plant in Sari, Iran and tannin that was purchased from Merck, Germany. Thickness of panels was controlled by

Table 1 Experimental design

Tablica 1. Osnovni parametri eksperimenta

Panel type <i>Vrsta ploče</i>	WS wt. %	MF wt. %	$T_{10\%}$ MF wt. %	$T_{20\%}$ MF wt. %	$T_{10\%}$ UF wt. %	$T_{20\%}$ UF wt. %	Temperature press <i>Temperatura prešanja</i> °C
A ₁₇₀	90	10	–	–	–	–	170
A ₁₈₀	90	10	–	–	–	–	180
B ₁₇₀	90	–	10	–	–	–	170
B ₁₈₀	90	–	10	–	–	–	180
C ₁₇₀	90	–	–	10	–	–	170
C ₁₈₀	90	–	–	10	–	–	180
D ₁₇₀	90	–	–	–	10	–	170
D ₁₈₀	90	–	–	–	10	–	180
E ₁₇₀	90	–	–	–	–	10	170
E ₁₈₀	90	–	–	–	–	10	180

Table 2 Production parameters of MDFs

Tablica 2. Parametri proizvodnje MDF ploča

Parameter / <i>Obilježje</i>	Value <i>Vrijednost</i>
Pressing time / <i>Vrijeme prešanja</i> , min	7
Peak pressure / <i>Tlak prešanja</i> , kg/mm ²	35
Thickness / <i>Debljina</i> , mm	12
Dimensions / <i>Dimenzije</i> , mm	420×420
Number of panels for each type <i>Broj proizvedenih ploča za svaki tip ploče</i>	3

stop bars and panels target density was 0.7 g/cm³. Three panels were produced for each group. The experimental design is shown in Table 1.

The dimensions of the produced panels were 42 × 42 × 1.2 cm. The produced MDF panels were conditioned at 20 °C and 65 % relative humidity to reach moisture content of about 12 % before trimming to final dimension of 40×40×1.2 cm. The panel production parameters are also displayed in Table 2.

Some mechanical properties: modulus of rupture (*MOR*), modulus of elasticity (*MOE*) (EN 310), internal bond strength (*IB*) (EN 319), and physical properties: thickness swelling (*TS*) and water absorption (*WA*) were determined for the produced fiberboards (EN 317). The average of 10 and 20 measurements were reported for mechanical and physical properties, respectively. The formaldehyde emission of the samples was measured according to the EN 717-3 standard. Six sheets of test pieces (with dimensions of 12 × 25 × 25 mm) were clamped 40 mm above water level using a plastic support in a 500-ml polyethylene bottle containing 50 ml of distilled water. The flask containing the specimens was loaded in the oven at 40 °C for 180 min. Formaldehyde released from the specimens was absorbed by the distilled water, which was subsequently used as the sample solution. The formaldehyde concentration in the sample solution was determined using acetylacetone–ammonium acetate solution and the acetylacetone method, with colorimetric detection at 412 nm. The data obtained was statistically analyzed using analysis of variance (ANOVA). Duncan’s group- ing was also included.

3 RESULTS AND DISCUSSION

3. REZULTATI I RASPRAVA

Table 3 shows the results of mechanical properties of produced MDF panels. The highest *MOR* (27.12 N/mm²) and *MOE* (2685 N/mm²) values were measured for fiberboard produced employing neat MF resin. Besides, the lowest *MOR* (22.78 N/mm²) and *MOE* (2285 N/mm²) values were determined for panels type E, including 20 % tannin in UF resin. The result indicated that, with increasing tannin content in the mixture, the *MOR* and *MOE* values of fiberboards significantly decreased. Statistical analysis found some significant differences ($P < 0.01$) between some group means for *MOR* and *MOE* values. Significant differences between groups were determined individually for these values by Duncan’s multiple comparison tests. The results of Duncan’s multiple range tests are shown in Table 3, denoted by letters. Depending on the amount of tannin content in the panels, average decreases in *MOR* values varied from 1.4 % to 16.1 % as compared to the average of the panel type A. All panel types showed significant differences ($P < 0.01$) from each other in their *MOR* properties. *MOE* values showed relatively similar trends to those of *MOR*. The average *MOE* values of the panels with tannin content decreased from 10.4 % to 14.9 % as compared to values of the panels made with unmodified MF resin (panel type A). With the exception of panel types B and C, as well as D and E, other panel types showed statistically significant differences ($P < 0.01$) from each other in their *MOE* properties. Addition of tannin content to panels had more adverse effect on *MOR* than on *MOE* values. The average *MOR* values of all panel types met the minimum requirement (22.78 MPa) of EN 622–5 for general-purpose MDF panels for use in dry conditions. However, only average *MOE* values of the panel type A met the minimum requirements for general purpose MDF (2500 MPa) of EN 622–5.

IB values of the experimental panels ranged from 0.53 to 0.70 N/mm². The highest *IB* value was observed for panel A, while the lowest was recorded for E type panel. *IB* values decreased with the increasing tannin content in the panels. The average decreases of

Table 3 Mechanical properties of MDF panels made from UF/tannin and MF/tannin and wood fibers and test results of ANOVA and Duncan's mean separation**Tablica 3.** Mehanička svojstva MDF ploča proizvedenih od smjese UF/tanina odnosno MF/tanina i drvnih vlakana te rezultati testa ANOVA i Duncanova testa srednjih vrijednosti

Mechanical properties <i>Mehaničko svojstvo</i>	Panel type <i>Vrsta ploče</i>	Mean ^a <i>Srednja vrijednost</i>	Std. deviation <i>Standardna devijacija</i>	Std. error <i>Standardna pogreška</i>	X_{Min}^b	X_{Max}^c	P^d
MOR (N/mm ²)	A	27.12 ^s	0.289	0.130	25.10	24.92	*
	B	26.75 ^u	0.235	0.089	24.60	24.65	*
	C	25.19 ^t	0.119	0.041	23.80	23.54	*
	D	24.45 ^v	0.127	0.052	23.02	22.41	*
	E	22.78 ^w	0.228	0.088	20.90	21.90	*
MOE (N/mm ²)	A	2685 ^s	34.995	13.975	2480.00	2580.00	*
	B	2407 ^u	13.754	5.545	2468.00	25200.00	*
	C	2395 ^u	33.389	13.980	2414.00	2490.00	*
	D	2412 ^t	27.003	9.995	2348.00	2385.00	*
	E	2285 ^t	11.680	4.855	2292.00	2376.00	*
IB (N/mm ²)	A	0.70 ^s	0.028	0.011	0.59	0.66	*
	B	0.68 ^s	0.012	0.005	0.58	0.63	*
	C	0.62 ^u	0.018	0.007	0.54	0.60	*
	D	0.55 ^u	0.016	0.006	0.52	0.58	*
	E	0.53 ^t	0.014	0.005	0.50	0.53	*

^a Mean values are the average of 10 specimens. / *Srednje vrijednosti dobivene su na temelju mjerenja provedenih na 10 uzoraka.* ^b Minimum value / *minimalna vrijednost;* ^c Maximum value / *maksimalna vrijednost;*

^d Significance level of 0.01 (for ANOVA) / *razina značajnosti 0,01 (za test ANOVA).*

^{s,u,t,v,w} Values having the same letter are not significantly different (Duncan test). / *Vrijednosti koje imaju isto slovo nisu statistički signifikantno različite.*

IB value, upon increasing the tannin content, were 12.9 % to 24.3 % as compared to values of the panels made using neat MF resin (panel type A). With the exception of panel types A and B, as well as C and D, other panel types showed statistically significant differences ($P < 0.01$) from each other in their IB properties. Out of the produced panels, only panel types A, B and C met the IB requirement (0.60 MPa) of EN 622–5 for general-purpose MDF panels for use in dry conditions.

The strength properties of wood products depend on many factors such as physical and mechanical properties as well as compact ratio of employed wood species, the configuration (orientation) and interfacial adhesion or interphase quality (Aydin *et al.*, 2006; Ciannamea *et al.*, 2010; Papadopoulos and Hague, 2003). Employed resin (bonder) plays a crucial role in wood based panels so that the quality of bonding, and hence the properties of the resulting materials, are dominantly determined by the type and quality of the bonder. There is close relationship between the ratio of urea and formaldehyde so that, with decreasing formaldehyde below a certain point, mechanical strength properties of the resulting UF resin will be seriously affected (Copur *et al.*, 2008). Besides, chemical composition (lignin, cellulose and hemicellulose contents) of wood species has a strong influence on mechanical properties (Habibi *et al.*, 2008) so that cellulose as well as hemicellulose contents have an impressive effect on surface wettability and hence adhesion (Aghakhani *et al.*, 2013).

Unidirectional cellulose microfibrils constitute the reinforcing elements in the matrix blend of hemicellulose and lignin (Dunky and Pizzi, 2002). The effect of press temperature on mechanical properties of the resulting MDFs was not significant and this is why

the result has not been included in the result and discussion part. The reason can be due to narrow temperature range used in this study.

Similar results have been published by different authors, who used various bio wastes (Aydin *et al.*, 2006; Nemli and Colakoglu, 2005; Nemli *et al.*, 2009). The results of ANOVA and Duncan's mean separation test for TS and WA of panels, made using the mixture of different adhesive formulations and wood fibers for 2 and 24 h water immersion times, are given in Table 4.

Adding tannin into MDF improved water repellency. With increasing tannin in the panels, the TS and WA values decreased from 3.3 % to 33.1 % and 4.5 % to 21.8 % for 2 h water immersion time, respectively. Likewise, for 24 h water immersion time, these figures were 3.4 % to 14.7 % and 2.1 % to 17.1 %, respectively. The average TS values of all panel types showed significant difference ($P < 0.01$) from each other with the exception of panels D and E in TS after 2 h water immersion time. Besides, average WA values of all panel types showed significant difference ($P < 0.01$) from each other after 2 and 24 h water immersion time. TS values of all panels did not meet maximum property requirements of 15 % for 24h water immersion for general purpose MDF panels specified by EN 622–5 (2005) standard.

The water resistance properties of wood based composites are affected by both water uptake capacity of lignocellulosic raw material and adhesive bonding quality (Roumeli, *et al.*, 2010). The effect of press temperature on physical properties of the resulting MDFs was not significant and this is why the result has not been included in the result and discussion part. The reason can be due to narrow temperature range used in this

Table 4 Test results of thickness swelling (TS) of MDF panels produced from UF/tannin and MF/tannin and wood fibers and of ANOVA and Duncan's mean separation

Tablica 4. Bubrenje u debljinu MDF ploča proizvedenih od smjese UF/tanina odnosno MF/tanina i drvnih vlakana te rezultati testa ANOVA i Duncanova testa srednjih vrijednosti

Physical properties <i>Fizikalno svojstvo</i>	Panel type <i>Vrsta ploče</i>	Soaking time <i>Vrijeme potapanja min</i>	Mean ^a <i>Srednja vrijednost</i>	Std. deviation <i>Standardna devijacija</i>	Std. error <i>Standardna pogreška</i>	X_{Min}^b	X_{Max}^c	p^d
Thickness swelling <i>bubrenje u debljinu (TS)</i>	A	2	12.01 ^s	0.201	0.089	10.26	12.40	*
	B	2	11.63 ^u	0.525	0.195	10.20	11.65	*
	C	2	8.85 ^t	0.354	0.112	10.06	11.05	*
	D	2	8.50 ^v	0.343	0.085	9.54	10.62	*
	E	2	8.10 ^v	0.338	0.124	9.65	9.58	*
	A	24	21.25 ^s	0.404	0.123	20.35	21.80	*
	B	24	20.52 ^s	0.352	0.135	19.67	21.30	*
	C	24	19.83 ^u	0.235	0.068	18.54	20.52	*
	D	24	19.02 ^t	0.429	0.163	18.33	19.30	*
	E	24	18.12 ^v	0.295	0.095	16.63	18.30	*
Water absorption <i>upijanje vode (WA)</i>	A	2	25.80 ^s	0.358	0.224	25.60	26.16	*
	B	2	24.65 ^u	0.457	0.186	24.02	25.12	*
	C	2	23.62 ^t	0.364	0.224	23.41	23.52	*
	D	2	21.36 ^v	0.428	0.163	21.39	22.36	*
	E	2	20.18 ^w	0.309	0.096	20.01	20.56	*
	A	24	34.41 ^s	0.421	0.136	33.56	34.50	*
	B	24	33.68 ^u	0.402	0.128	31.02	33.22	*
	C	24	31.48 ^t	0.708	0.298	30.54	32.52	*
	D	24	29.36 ^v	0.375	0.104	28.37	29.00	*
	E	24	28.54 ^w	0.319	0.095	28.09	28.45	*

^a Mean values are the average of 10 specimens. / *Srednje vrijednosti dobivene su na temelju mjerenja provedenih na 20 uzoraka.* ^b Minimum value / *minimalna vrijednost;* ^c Maximum value / *maksimalna vrijednost;* ^d Significance level of 0.01 (for ANOVA) / *razina značajnosti 0,01 (za test ANOVA).*

^{s,u,t,v,w} Values having the same letter are not significantly different (Duncan test). / *Vrijednosti koje imaju isto slovo nisu statistički signifikantno različite.*

study. Similar results have been reported for different bio wastes (Aydin, *et al.*, 2006; Kim and Kim, 2005).

3.1 Formaldehyde emission

3.1. Emisija formaldehida

The addition of Tannin into the panels significantly decreased formaldehyde emission value (Fig. 3.). Depending on addition of tannin in the panels, the formaldehyde emission values ranged from 15 % to 35% lower than the panels made with neat MF resin. The minimum value of formaldehyde gas emission was measured for panels made with 20 % tannin in melamine formaldehyde resin. To be specific, panel type C had the lowest formaldehyde emission value with 6.5 mg/100 g (35 % less than panel type A), followed by panel type B (8.5 %), panel type D (9 %), panel type E (9.3 %), and finally panel type A (10.2 mg/100 g) (Fig. 3.).

The results showed that the free-formaldehyde emission values of MDF panel types (E and C) for E1 class met the requirement of maximum 8 mg/100 g (Aydin *et al.*, 2006). The decrease in formaldehyde emission of MDF panels with increasing tannin can be attributed to the scavenging effect of tannin and its chemical bonds with formaldehyde (Aydin *et al.*, 2006). The similar results were reported in previous studies (Ayrilmis *et al.*, 2009; Nemli and Aydin, 2007). They reported that the decrease in formaldehyde emission values in the panels may be due to high amounts of polyphenolic extractives in bark, especially tannins (Lei *et al.*, 2009).

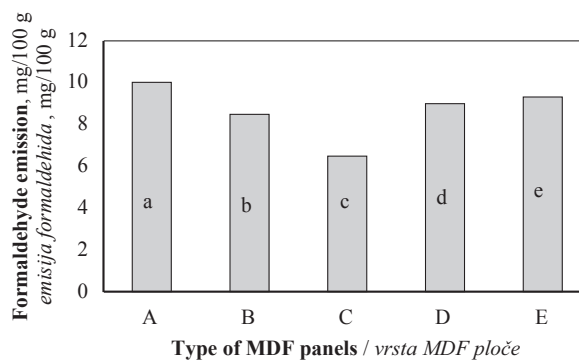


Figure 3 Percent decrease in average value of formaldehyde emission of panel types

Slika 3. Postotno smanjenje prosječne emisije formaldehida za sve vrste proizvedenih ploča

et al., 2009). It was found that incorporation of mimosa bark particle greatly reduces formaldehyde emission of particleboards. Post-treatment methods, aimed at decreasing and minimizing formaldehyde release, are based on compounds like ammonia, ammonium salts, or urea (Nemli and Aydin, 2007). Another effective way to reduce formaldehyde release is the addition of formaldehyde-binding substances ("scavengers") to the resin or to wood particles (Aydin *et al.*, 2006). Ammonium chloride acts as an acid catalyst of the curing reaction and as a formaldehyde scavenger (Kim, 2009). Coating panel surfaces with decorative overlays are to eliminate the

release of formaldehyde (Lei *et al.*, 2009). The environmentally friendly tannin–formaldehyde resins are among the cheapest binders and cause low-formaldehyde emission (Ayrilmis *et al.*, 2009). Besides, processing conditions of wood based panels have a serious impact on relative formaldehyde emission so that the higher the press cycle time, the lower the formaldehyde emission at service (Aghakhani, 2013). It is to be noted that using nanoparticles, such as nano-SiO₂ and nano-clay, due to their strong absorbability and high barrier properties (shielding effect) as formaldehyde catchers, will soon gain much attention (Kim, 2009; Papadopoulos and Hague, 2003).

4 CONCLUSION

4. ZAKLJUČAK

This study revealed that value-added MDF panels containing tannin can be considered as an alternative solution for decreasing carcinogenic gas of formaldehyde from wood based panels and for moving towards green chemistry. The incorporation of tannin greatly improved the formaldehyde emission of the panels; however, mechanical properties decreased with increasing tannin ratio. Using silane, dewaxing or raw material washing, the properties of wood based panels could be improved. Decreasing formaldehyde emission from MDFs containing tannin should be considered for furniture materials used indoor and this would be significant movement towards green chemistry and products.

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