FORMALDEHYDE BARRIER EFFICIENCY OF MELAMINE IMPREGNATED PAPER AND MELAMINE EDGE-BANDING TAPE SURFACED PARTICLEBOARD AND MDF UČINKOVITOST PAPIRJA, IMPREGNIRANEGA Z MELAMINSKO SMOLO, IN MELAMINSKEGA ROBNEGA TRAKU PRI OBLAGANJU IVERNIH PLOŠČ IN MDF-PLOŠČ Z VIDIKA ZMANJŠANJA EMISIJE FORMALDEHIDA

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ABSTRACT

Particleboard and MDF are the most usable materials for furniture production, where emission of formaldehyde should be as low as possible. In order to improve aesthetic properties of particleboard and MDF they are covered with appropriate surfacing material. In most cases, melamine impregnated paper is used for face layers. But covering faces should be accompanied with covered edges, as due to low density of core layer formaldehyde much more easily emits from edges than from faces. The presented research shows differences in formaldehyde emission rate at unsurfaced and surfaced MDF and particleboard. For face layer, melamine impregnated papers with grammages of 130 g/m² and 200 g/m² were used, while for covering edges, melamine edge-banding tape was applied. In the research it was determined that higher barrier effect was achieved when impregnated paper with a grammage of 200 g/m² was used. When surface was completely covered with impregnated paper and melamine edge-band tape, formaldehyde emission was lowered by between 83 % and 94 %.

Key words: particleboard, MDF, surfacing, melamine impregnated paper, melamine edge band-tape, formaldehyde emission

IZVLEČEK

Iverne in MDF-plošče so najpogosteje uporabljen material za izdelavo pohištva, kjer se zahteva čim nižja emisija formaldehida. MDF- in iverne plošče se s ciljem doseganja želenih estetskih karakteristik površinsko obdelajo z ustreznimi materiali. Največ-krat so to papirji, impregnirani z melaminsko smolo. Samo oblaganje ploskve pa z vidika zmanjšanja emisije formaldehida ni zadostno, saj formaldehid bistveno laže prehaja skozi rob, kar posledično pomeni, da je treba obdelati oz. zapreti tudi robove. V predstavljeni raziskavi smo iverne in MDF- plošče obložili z impregniranim papirjem gramature 130 g/m² in 200 g/m², robove pa zaprli z melaminskim robnim trakom. Efektivnejša zaščita proti emisiji formaldehida je bila dosežena z uporabo impregniranega papirja gramature 200 g/m². V primeru, da so bili zaprti tako robovi plošč kakor tudi ploskve, se je emisija formaldehida zmanjšala med 83 % in 94 %.

Ključne besede: iverna plošča, MDF, oblaganje, papir impregniran z melaminsko smolo, melaminski robni trak, emisija formaldehida

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1 INTRODUCTION

1 UVOD

Formaldehyde is one of most criticized property of particle and fibre-based wood based panels. Woodbased panel producer and with them related resin producers are constantly looking for the solutions to improve (lower) formaldehyde emission from panels, especially those used for furniture. One of the most widely used wood-based panels for furniture are particleboards (PB). The usability of raw (unsurfaced) particleboards in furniture application is rare due to their unappealing look and less resistant surface especially against water/moisture and mechanical damages. To improve aesthetic look, resistance towards water/moisture, scratch and abrasion resistance boards are surfaced/laminated/overlaid. (Groah et al., 1984; Nemli and Çolakoğlu, 2005; Nemli et al., 2005; Nemli and Hiziroglu, 2009; Istek et al., 2010; Bardak et al. 2011). Surfacing/laminating/overlaying describes a process where impregnated paper, foil or laminate type of covering material is applied on the wood-based panel surface using pressure and elevated temperature. In addition to that, also edges are mostly protected by using different edging-banding materials like ABS (Acrylonitrile Butadien Styrene), PVC, melamine, solid wood or veneer (Tankut and Tankut, 2010).

Although surfacing of PB, even just with very thin impregnated paper (thickness < 0.3 mm; paper grammage or base weight between 45 g/m² and 200 g/m²), lowers the formaldehyde emission (Liu et al., 2015), the issues related to formaldehyde emissions are still present. Despite the fact that over the last few decade's producers of wood-based panels significantly lowered formaldehyde emission (Salthammer et al. 2010), nowadays we are facing different challenge related to the indoor formaldehyde concentrations. Today, building practice as well as users demand high-energy efficient buildings, which is related to the decreased air exchange rate inside the building. Such low air exchange activity enables accumulation of formaldehyde that emits from materials. The concentration of formaldehyde in indoor air, although emitting from materials with low formaldehyde concentration (like PB or MDF, where concentration of formaldehyde is lower than 0.1 ppm as permitted by EN 312 or EN 622-1) can accumulate to such level/concentration that could cause eye and respiratory irritation.

As mentioned before, surfacing of PB and MDF can lower formaldehyde emission, but closing the surface is not enough. As was shown in research conducted by Grigoriou (1987), it is important to seal edges as well. Although the edge apparent surface area is lower than the face surface area, the formaldehyde emission from edges is higher than that from or through face layer. According to Barry and Corneau (2006), 96 % reduction of formaldehyde emission could be achieved if panels were well surfaced and edges covered. Effect of panel area to volume ratio on release of volatile organic compounds at surfaced particleboards were also investigated by Yali et al. (2018). It was determined that increase in panel area to volume ratio led to the increase in volatile organic compounds concentration.

Today's furniture producer usually are sealing only visible edges in order to save money and make cheaper products, hence leaving at least two edges open and with that enabling easy emission of formaldehyde from board to indoor air. Easy emission of formaldehyde from panels is further enhanced due drilling different holes. Some of them are used for shelf holders, but the majority of them are left open what enables, despite covered face, the emission of formaldehyde.

The aim of this work is to show the efficiency of creating a formaldehyde emission barrier by applying melamine impregnated paper and melamine based edging-banding material.

2 MATERIALS AND METHODS

2 MATERIALI IN METODE DELA

For the purpose of investigation furniture grade, three-layer particleboard (PB), thickness 18 mm and density 650 kg/m³, and MDF, thickness 12 mm and density 760 kg/m³, available on Slovenian market (contributed by the firm Starman) were used. The impregnated papers used in the research were contributed by Melamin Kočevje, Slovenia. Two types of impregnated paper were used, specifically paper with grammages of 130 g/m² (IP130) and 200 g/m² (IP200). Melamine based edge-banding tape available on Slovenian market was used (contributed by the firm Blažič, robni trakovi, d.o.o., Slovenia).

After seven-day exposure to normal climate condition (temperature 20±1°C and relative air humid-

Table 1: Overview of testing material

Preglednica 1: Pregled preskušancev

Panel / <i>Plošča</i>	Type / Vrsta	Surface / Ploskev	Edge / Rob	
A1	PB	-	-	
A2	PB	-	Al	
A3	PB	Al	-	
B1	MDF	-	-	
B2	MDF	-	Al	
B3	MDF	Al	-	
A1/130	PB	IP130	-	
A2/130	PB	IP130	Al	
A5/130	PB	IP130	М	
A1/200	PB	IP200	-	
A2/200	PB	IP200	Al	
A5/200	PB	IP200	Μ	
B1/200	MDF	IP200	-	
B2/200	MDF	IP200	Al	
B5/200	MDF	IP200	М	

ity 65±5 %), 500×500 mm² panels were surfaced with melamine impregnated paper. The surfacing was done in a laboratory press, where upper heating plate was set at the temperature of 185°C and lower heating plate at 180°C. The pressure was 3 N/mm², while pressing time was 30 seconds. After pressing, the boards were cooled at room temperature for 24 hours, followed by cutting to the sample size of 400×50 mm². On some samples, melamine based edge-banding tape was applied (Table 1). For the application of melamine edge-banding tape, hot-melt adhesive was used. Until measurement, samples were placed in airtight plastic bag and stored at normal climate conditions.

The determination of formaldehyde emission was done according to SIST EN 717-2 (gas analysis method). The samples with dimensions $400 \times 50 \text{ mm}^2$ were placed in a chamber that was heated to the temperature of $60 \pm 0.5^{\circ}$ C. The level of humidity in the chamber was 2 ± 1 % and the air flow into the chamber was 60 ± 3 l/h. Four sets of wash bottles containing from 30 to 40 mL of distilled water were connected to the chamber. Formaldehyde emission was measured for 4 hours at hourly intervals. After 4 hours the water solution from wash bottles was transferred into a 250 mL volumetric flask to which distilled water was added to the indicated mark. The water solution (10 mL) was transferred from the volumetric flask to another flask to which we added 10 mL of acetyl acetone and 10 mL of ammonium acetate solution. The flask was stoppered, shaken and placed in a water bath with temperature 40°C for 15 minutes. The solution was then placed in a dark chamber for one hour. The absorption of the solution was determined using a spectrophotometer at 412 nm. Formaldehyde emission rate (FER) was calculated according to equations 1 and 2.

$$FER_i = (A_s - A_b) \times f \times V \tag{1}$$

where:

FER_i is the formaldehyde content of the solution from each hour [mg/h]

Table 2: Formaldehyde emission at reference conditions(raw panel, surface or edge sealed with alluminium selfad-
hesive tape)

- i is the first, second, third or fourth hour
- A_s is the absorbance of the solution from the wash bottles (sample)
- A_b is the absorbance of distilled water
- f is the slope of the calibration curve for standard formaldehyde solution, in milligrams per millilitre
- V is the volume of the volumetric flask [mL] (250 mL)

$$FER_m = \frac{G_1 + G_2 + G_3 + G_4}{4} \tag{2}$$

where:

 FER_m is the average formaldehyde emission rate value of test piece [mg/h]

The barrier efficiency of surface closing was calculated by equation 3:

$$BE_{HCHO} = 1 - \frac{Surfaced panel emission rate}{Emission rate at group reference}$$
(3)

3 RESULTS AND DISCUSSION 3 REZULTATI IN RAZPRAVA

Exposed surface plays important role when formaldehyde emission is considered (Table 2).

The application of impregnated paper and edgebanding material significantly lowers formaldehyde emission (Figures 1 and 2).

The barrier effect of impregnated paper depends on the grammage of the paper. Higher barrier effect (at PB) concerned paper with higher grammage (0.88 compared to 0.69). The impregnated paper with higher grammage is more effective than formaldehyde barrier due to its higher thickness (0.15 mm to 0.2 mm compared to 0.1 mm to 0.12 mm) and more adhesive (around 120 g/m² compared to 80 g/m²), hence it seals the faces more efficiently. Although we surfaced MDF board with impregnated paper with a grammage of 200, the barrier efficiency was not as high as observed at PB. The barrier effect at board B2/200 (MDF sur-

Preglednica 2: Emisija formaldehida v referenčnih razmerah (surova plošča, ploskev oz. rob zaprta s samolepilnim aluminijastim trakom)

Panel / <i>Plošča</i>	Type / <i>Vrsta</i>	Face / <i>Ploskev</i>	Edge / Rob	Emission surface Površina emitiranja	Formaldehyde emission rate Stopnja emisije formaldehida [mg/h]
A1	PB	-	-	All	0,1831
A2	PB	-	AI	Face	0,1463
A3	PB	AI	-	Edge	0,1255
B1	MDF	-	-	All	0,2652
B2	MDF	-	AI	Face	0,2359
B3	MDF	AI	-	Edge	0,1542

faced with IP200) was 0.64. Possible reason for the lower efficiency lies in higher formaldehyde emission observed at MDF.

The reason for formaldehyde barrier effect at thin melamine impregnated paper (thickness less than 0.3 mm) is related firstly due to the creation of additional



Fig. 1: Formaldehyde emission of surfaced PB with IP130 (A2/130) and IP200 (A2/200), MDF with IP200 (B2/200) and closed edges with Al tape; blue line represents the reference value for the PB group – 0.1463 mg/h, and red dashed line for MDF group – 0.2359 mg/h

Slika 1: Emisija formaldehida pri PB, obloženi z IP130 (A2/130) in IP200 (A2/200), ter MDF, obloženi z IP200 (B2/200) in z Al-trakom zaprtimi robovi; modra črta predstavlja referenčno vrednost za PB skupino – 0.1463 mg/h, rdeča črtkana črta pa za MDF-skupino – 0.2359 mg/h



Fig. 2: Formaldehyde emission of surfaced PB and MDf with opened (A2/130, A2/200 and B2/200) and closed edges with melamine based edge-band closed edges (A5/130, A5/200 and B5/200)

Slika 2: Emisija formaldehida pri obloženi PB in MDF z odprtimi robovi (A2/130, A2/200 in B2/200) in z melaminskim robnim trakom zaprtimi robovi (A5/130, A5/200 in B5/200) diffusion resistant layer in addition to already existing diffusion resistance face layer (due higher density compared to core layer). Second reason is due to the pressing conditions. Panels are pressed at a high temperature (usually higher than 180°C), which causes heat and mass flow towards the core layer, and with that also migration of formaldehyde from face into core layer, which results in the accumulation of formaldehyde in core layer. Similar effect was also determined by Grigoriou (1987). During pressing, formaldehyde also emits from panel through edges (with moisture evaporation) and after opening of the press. This was also determined during the experiment. The unsurfaced (raw) particleboard was exposed to same conditions as occurring during surfacing (upper heating plate 185°C, lower heating plate 180°C, pressing pressure 3 N/mm², pressing time 30 seconds). The FER for "tempered" panel was 0.1183 mg/h (compared to raw particleboard FER which was 0.1831).

As could be seen by comparing the results in Table 2 and Figure 1, panel surfacing is an effective way to reduce formaldehyde emission, but in order to prevent easy emission from the panels, edges also needs to be closed. Closing edges lowers formaldehyde emission by at least 40 % (Figure 2).

The formaldehyde emission rate from panels with opened edges was between 0.0665 and 0.0774 mg/h but when edges were closed with melamine edge-band tape, FER was lowered by between 42 % and 59 %. Despite the fact that surface of the faces is much higher than edge surface (0.04 m² compared to 0.0162 m² at

PB respectively 0.0108 m² at MDF) the barrier effect of edge-band is high. The reason lies in the diffusion resistance. The density of core layer is lower than surface layer, hence the formaldehyde diffusion resistance of core layer is lower as well. Formaldehyde emits from panel through the path that offers least resistance and since faces present highest diffusion resistance (higher density) formaldehyde emits from edges, but when edges are closed (melamine edge-band) it can be assumed that the majority of formaldehyde emission occurs through semipermeable décor paper.

In Figures 3 and 4, the comparison of FER values of unsurfaced and surfaced panels is shown.

In order to achieve the highest possible barrier effect, all surfaced boards need to be covered. In case some surfaces like edges or even drilled holes are left open, then higher formaldehyde emissions at the customers' end can occur.

4 CONCLUSIONS

4 ZAKLJUČKI

The results of our experiment pointed at the importance of closing the surfaces of particleboard and MDF in order to lower the formaldehyde emission.

The highest barrier effect was determined when melamine impregnated paper with a grammage of 200 g/m^2 was used.

Closing surface decreased formaldehyde emission by 58 % to 71 %. Closing edges additionally decreased formaldehyde emission by 12 % to 30 %.





Slika 3: FER in BE_{HCHO} vrednosti surovih in obloženih PB



Fig. 4: FER and $\mathsf{BE}_{_{\mathsf{HCHO}}}$ values of unsurfaced and surfaced MDF

Slika 4: FER in BE_{HCHO} vrednosti surovih in obloženih MDF

5 POVZETEK

5 SUMMARY

Iverne plošče in MDF-plošče so najpogostejši materiali, ki se uporabljajo za izdelavo pohištva, kjer je poleg dobrih mehanskih in sorpcijskih lastnosti pomembna tudi emisija formaldehida. Čeprav so se vrednosti emisije formaldehida od leta 1980 občutno zmanjšale (Salthammer et al. 2010), pa so zaradi načina gradnje (majhna cirkulacija zraka) nastale težave zaradi akumulacije formaldehida v bivalnih prostorih. Za izdelavo pohištva so površinsko neobdelane plošče neprivlačne, zato jih proizvajalci ustrezno površinsko obdelajo največkrat s papirji, impregniranimi z melaminsko smolo. Naloga teh papirjev je, poleg estetske/ dekorativne, tudi povečanje odpornosti proti vodi, mehanskim poškodbam, kakor tudi izboljšanje nekaterih mehanskih lastnosti (Groah et al., 1984; Nemli and Çolakoğlu, 2005; Nemli et al., 2005; Nemli and Hiziroglu, 2009; Istek et al., 2010; Bardak et al. 2011). Z oblaganjem se zmanjša tudi emisija formaldehida (Liu et al., 2015). S ciljem čimbolj učinkovitega zmanjšanja emisije formaldehida pa je pomembno, da se zapro tudi robovi plošč (Grigoriou, 1987). Proizvajalci pohištva zaradi zmanjševanja stroškov zapirajo predvsem vidne robove, kar pomeni, da sta največkrat zaprta zgolj dva robova. Zaradi manjše gostote robov in posledično manjše difuzijske odpornosti pa formaldehid hitreje prehaja prek robov kakor prek ploskve (Grigoriou, 1987). Namen raziskave je prikazati učinkovitost oblaganja z impregniranim papirjem in melaminsko robno folijo z vidika zmanjšanja emisije formaldehida.

Za raziskavo smo uporabili 18 mm debelo trislojno iverno ploščo, namenjeno za izdelavo pohištva, in 12 mm debelo MDF-ploščo, prav tako namenjeno za izdelavo pohištva. Za oblaganje smo uporabili papir, impregniran z melaminsko smolo površinske teže 130 g/m² in 200 g/m² (Melamin Kočevje), za zapiranje robov smo uporabili melaminski robni trak (Blažič, robni trakovi, d.o.o.). Trakove smo prilepili s talilnim lepilom.

Emisijo formaldehida smo določili po plinski metodi (SIST EN 717-2), in sicer na preskušancih velikosti 400×50 mm². Stopnjo emitiranja formaldehida smo izračunali po enačbah 1 in 2.

$$FER_i = (A_s - A_b) \times f \times V \tag{1}$$

Kjer je:

FER_i urna stopnja emitiranja formaldehida [mg/h]

- i prva, druga in četrta ura vzorčenja
- A_s absorpcija preskušanca
- A_b absorpcija, določena pri destilirani vodi (slepa proba)
- f naklon kalibracijske krivulje

V volumen merilne bučke [mL] (250 mL)

$$FER_m = \frac{G_1 + G_2 + G_3 + G_4}{4} \tag{2}$$

kjer je:

FER_m povprečna stopnja emitiranja formaldehida [mg/h]

Preglednica 3: Stopnje emisije formaldehida in učinkovitost impregniranega papirja robnega traku **Table 3**: Formaldehyde emission rate and barrier effect ofimpregnated paper and edge-banding tape

Plošča	Vrsta	Ploskev	Rob	Površina emitiranja	FER [mg/h]	BE _{нсно}
A1	PB	-	-	Vsa	0,1831	1
A2	PB	-	AI	Ploskev	0,1463	0,31
A3	PB	AI	-	Rob	0,1255	0,31
B1	MDF	-	-	Vsa	0,2652	1
B2	MDF	-	AI	Ploskev	0,2359	0,11
B3	MDF	AI	-	Rob	0,1542	0,42
A1/130	PB	IP130	-	Vsa	0,0768	0,58
A5/130	PB	IP130	М	Ploskev	0,0318	0,83
A1/200	PB	IP200	-	Vsa	0,0665	0,64
A5/200	PB	IP200	М	Ploskev	0,0116	0,94
B1/200	MDF	IP200	-	Vsa	0,0774	0,71
B5/200	MDF	IP200	М	Ploskev	0,0452	0,83

Izračunali smo tudi učinkovitost impregniranega papirja in robnega traku z vidika zmanjšanja emisije formaldehida (enačba 3).

$$BE_{HCHO} = 1 - \frac{Stopnja \ emisije \ obložene \ pložče}{Stopnja \ emisije \ v \ referenčnih \ razmerah} \tag{3}$$

Stopnje emisije formaldehida in učinkovitost impregniranega papirja robnega traku so prikazani v preglednici 3, kjer smo kot osnovo za izračun vrednosti BE_{HCHO} uporabili FER-vrednosti surovih plošč.

Z zapiranjem ploskve in roba se emisija formaldehida občutno zmanjša. Pomembno vlogo ima zapiranje robov, saj formaldehid laže prehaja iz plošče prek ploskev z nižjo gostoto, kjer je tudi odpornost proti emitiranju formaldehida manjša, prav tako pa med samim stiskanjem (izdelavo plošč in oblaganjem) se formaldehid seli v sredico plošče, kjer posledično prihaja do akumulacije formaldehida. Če robovi plošč ostanejo odprti, prihaja posledično do hitrejšega emitiranja v okolico, kar pomeni večja koncentracija formaldehida v zraku. Ugotovili smo tudi, da je stopnja emitiranja formaldehida pri uporabi impregniranega papirja večje površinske teže manjša.

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