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Evaluation of Mechanical Properties and Formaldehyde Emission of Plywood Manufactured for Construction Applications

Ocjena mehaničkih svojstava i emisije formaldehida furnirske ploče proizvedene za primjenu u graditeljstvu

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ABSTRACT • *This study was aimed to evaluate some mechanical properties and formaldehyde emission of beech plywood panels manufactured from rotary cut veneers using phenol–formaldehyde (PF) resin. Six plywood panels of 5 plies with 15 mm thickness were manufactured. Modulus of rupture (MOR), modulus of elasticity (MOE) and formaldehyde emission values were determined. The values of MOR and MOE were higher than the EN requirements. Furthermore, the formaldehyde emission values measured by the European small-scale chamber (EN 717-1) and gas analysis (EN 717-2) methods were lower than the E1-emmison class. The results showed significant variations (P<0.001) among beech plywood panels, most of them caused by sample heterogeneity, and inconsistency of the values was related to the inter-panel variation. Moreover, a strongly positive correlation between the formaldehyde emission values measured by EN 717-1 and 717-2 methods was found (R^2 value of 0.87).*

Keywords: *mechanical properties, formaldehyde emission, beech plywood, European small-scale chamber, gas analysis.*

SAŽETAK • *Cilj istraživanja bio je procijeniti neka mehanička svojstva i emisiju formaldehida bukovih furnirskih ploča proizvedenih od ljuštenog furnira i fenol-formaldehidne (PF) smole. Proizvedeno je šest furnirskih ploča od pet slojeva furnira debljine 15 mm. Utvrđene su vrijednosti modula loma (MOR), modula elastičnosti (MOE) i emisija formaldehida. Vrijednosti MOR i MOE veće su od zahtjeva europskih normi. Nadalje, vrijednosti emisije formaldehida izmjerene metodom europske male komore (EN 717-1) i metodom plinske analize (EN 717-2) bile su niže od klase emisije E1. Rezultati su pokazali značajne razlike (P <0,001) među bukovim furnirskim pločama, a one su posljedica heterogenosti uzoraka. Nedosljednost izmjerenih vrijednosti povezana je i s varijacijama unutar*

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iste ploče. Također, utvrđena je jaka pozitivna korelacija između vrijednosti emisije formaldehida mjerene metodom EN 717-1 i metodom EN 717-2 (R^2 vrijednost iznosi 0,87).

Ključne riječi: *mehanička svojstva, emisija formaldehida, bukove furnirske ploče, europska mala komora, plinska analiza*

1 INTRODUCTION
1. UVOD

Formaldehyde is a potential human carcinogen and, due to its high risk level, it is classified differently than most other pollutants. Many building materials including plywood emit formaldehyde, which has the potential to affect health and comfort. In the previous studies, the emissions of formaldehyde from wood-based panels were determined using differing standard test methods (Que and Furuno, 2007; Salem *et al*., 2011a; Salem *et al.*, 2011b; Salem *et al*., 2012a, Böhm *et al*., 2012; Park *et al.,* 2010; Roffael *et al*., 2010; Risholm-Sundman and Wallin, 1999).

The formaldehyde emission from wood-based panels, including plywood and flooring materials, has received great attention from the general public as well as wood industries, ever since formaldehyde became known as a toxic air contaminant. In fact, the International Agency for Research on Cancer (IARC), a part of the World Health Organization (WHO), reclassified formaldehyde from 'probably carcinogenic to humans (Group 2A)' to 'carcinogenic to humans (Group 1)' in June 2004 (IARC, 2004). Nowadays, health and the environment constitute two key concerns of the 21st century (Lee *et al*., 2002).

Plywood is one of the main products that can be used as structural material and has traditionally played an important role in light frame construction. Plywood and other wood-based products are widely used in the manufacture of furniture, engineered flooring, housing and other industrial products (Böhm *et al*., 2012). The plywood panels are manufactured by gluing several wooden plies together in layers, which have perpendicular grain directions. The thickness of plies is generally from 2 to 3mm (Arriaga-Martitegui *et al*., 2008). Where there are no requirements with respect to the appearance of the material, lower-grade veneers for plywood may be suitable for structural purposes without compromising the function of structural members.

Plywood is a complex product and the bending properties as a strength class system are defined in EN 636 (2003) for quality control procedures (EN 636-1; EN 636-2; EN 636-3). Furthermore, the determination of all the mechanical properties of plywood requires a large amount of time-consuming testing for each different board composition. Additionally, the factors affecting physico-mechanical properties and formaldehyde emission from plywood panels and its products and have been studied (Sensogut *et al*., 2009; Aydin *et al*., 2006; Aydin and Colakoglu, 2007; Colak and Colakoglu, 2004; Martínez and Belanche, 2000).

Urea formaldehyde (UF) and phenol- formaldehyde (PF) resins are widely used as a major component in the production of building and furniture materials. PF resin is used to manufacture plywood for exterior applications because of its excellent water resistance. Plywood products used as an exterior product are suitable for moist and wet use, carrying and non-carrying construction elements. Phenolic-based compounds also tend to be more chemically stable and less susceptible to hydrolysis than UF. Both of these characteristics are beneficial to PF resin. Moreover, PF tends to be more chemically stable and less susceptible to hydrolysis than UF and is considered waterproof, while UF is not (Salem *et al*., 2011b; Dunky, 2005). PF resin has generally been the resin chosen by manufacturers of exterior grade structural panels. In addition, the C-C bonding in the PF resins was very stable against hydrolytic attack. The objectives of this study were to evaluate the mechanical properties of beech plywood manufactured for structural application and to check the formaldehyde emission according to the European standards.

2 MATERIALS AND METHODS
2. MATERIJAL I METODE

2.1 Production of plywood panels and samples
2.1. Proizvodnja furnirskih ploča i uzoraka

Wood logs of 60 cm in diameter from European beech (*Fagus sylvatica* L.) were steamed for 48 h before the production of veneer. Sheets of veneer with dimensions of 50 cm × 50 cm × 3.2 mm were produced with rotary cuts and randomly chosen from the main veneer sheet, and then dried at 110 °C. Subsequently, the veneers were conditioned in a climate chamber to 4-6 % moisture content. Plywood (5-ply) with a nominal thickness of 15 mm was produced from beech veneers using PF resin (46 % solid content). The manufactured plywood was classified as external structural component (EN 636-3, 2003). The properties and composition of PF resin used are presented in Table 1. For manufacturing plywood panels, the following parameters were used: hot press time, pressure and temperature (Table 2). Six replicate panels (p1, p2, p3, p4, p5 and p6) were manufactured, wrapped with polyethylene film and delivered to the laboratory of Timber Research and Development Institute in Prague, Czech Republic.

2.2 Measuring of mechanical properties and formaldehyde emission
2.2. Mjerenje mehaničkih svojstava i emisije formaldehida

Since, as previously discussed, the determination of all the mechanical properties of plywood requires considerable time and effort, and the number of possible variables that define a type of plywood are large for the same thickness, it is possible to use different lay-ups, even different species, giving different mechanical

Table 1 Properties and composition of phenol-formaldehyde adhesive resin used for plywood
Tablica 1. Svojstva i sastav fenol-formaldehidnog ljepila upotrijebljenog za proizvodnju ploča

Parameter / *Parametar*	**phenol-formaldehyde adhesive resin** *fenol-formaldehidno ljepilo*
Solid resin content, % *Sadržaj krute tvari, %*	46
Viscosity (mPa·s at 20 °C) *Viskozitet, (mPa·s at 20 °C)*	250–1050
pH at 20 °C	min. 11.5
Density / *Gustoća*	1210–1250 kg/m³
Foaming agent[a] *Sredstvo za pjenjenje*	Oxyethylene castor oil and mixture of non-ionic tensides *oksietilen ricinusovo ulje i mješavina neionskih tenzida*
Free formaldehyde *Slobodni formaldehid*	max 0.1 % najviše 0,1 %

[a] PF resin + foaming agent (PF resin / foaming agent - 100:1) with density (PF resin + foaming agent): 750-850 kg/m³ / *PF smola + sredstvo za pjenjenje (PF smola / sredstvo za pjenjenje – 100:1) gustoće (PF smola + sredstvo za pjenjenje): 750-850 kg/m³.*

Table 2 Pressing conditions of plywood bonded with phenol-formaldehyde adhesive resin
Tablica 2. Uvjeti prešanja furnirskih ploča izrađenih fenol-formaldehidnim ljepilom

Parameter / *Parametar*	**Value** *Vrijednost*
Specific pressure / *specifični tlak*	1.5 MPa
Pressing temperature / *temperature prešanja*	118-124 °C
Pressing time / *vrijeme prešanja*	60 s / 1mm[a]
Veneer moisture / *sadržaj vode u furniru*	5±2 %
Veneer thickness / *debljina furnira*	≤ 3.2 mm
Adhesive spread / *nanos ljepila*	150 g/m²[b]

[a] Basic curing time for PF adhesive is 3 minutes and for the heating of 1 mm veneer thickness 1 minute. / *Osnovno je vrijeme stvrdnjavanja za PF ljepila 3 minute, a za zagrijavanje furnira debljine 1 mm iznosi 1 minutu.*
[b] Approximately 150 g/m² adhesive mixture was spread on single surfaces of veneers using a gluing machine. / *Na jednu površinu furnira prosječno je naneseno 150 g/m² smjese ljepila nanijeto je primjenom uređaja za nanos ljepila.*

properties (Arriaga-Martitegui *et al*., 2008). Therefore, in the present study the bending properties; modulus of rupture (MOR) and modulus of elasticity (MOE) were determined in accordance with EN 636-3 (2003) and EN 13986 (2004) standards.

Six samples from each panel of beech plywood, 15 mm thick, were prepared in accordance with the standards EN 636-3 and EN 13986 to measure the mechanical properties (MOR and MOE). These standards were designed for testing the requirements of plywood as well as wood-based panels. Additionally, six samples, representing randomly distributed portions of an entire panel, were edge–sealed with aluminum tape and conditioned at 23 °C ± 0.5 °C and relative humidity of 50% ± 5% (RH) for 10 days before the analysis of formaldehyde emission with gas analysis method (EN 717-2, 1994). Furthermore, formaldehyde emission was measured from another six samples by the European small-scale chamber method (EN 717-1, 2004), where the measuring was done after removing the polyethylene film.

In the referenced method (EN 717-1), two test pieces with the dimensions of 0.2 m × 0.28 m × 15 mm and a total area of 0.225 m² were used for the measuring of formaldehyde emission. The loading factor was 1 m²/m³ (1.5 m open edge/m²). The temperature and RH were 23 ± 0.5 °C and 45 ± 3 %, respectively. The values after 2 to 4 weeks of testing are given as the steady-state emission values. The E1-emission class is ≤ 0.1 ppm (0.124 mg/m³).

In the factory quality control method (EN 717-2), an edge-sealed test piece with self-adhesive aluminum tap of 400 mm × 50 mm × 15 mm was placed in a 4 L chamber with controlled temperature (60 ± 0.5 °C), relative humidity (RH ≤ 3%), airflow (60 ± 3 L/h) and pressure. The measuring of formaldehyde was repeated (six samples from each panel) for better homogeneity of the results. The E1-emission class was ≤ 3.5 mg/m² h. The emitted formaldehyde from both methods was absorbed in water and determined photometrically by the acetylacetone method (Nash, 1953).

2.3 Statistical analysis
2.3. Statistička analiza

The results of mechanical properties (MOR and MOE) and formaldehyde emission values were statistically analyzed using the SAS version 8.2 (2001) in a completely randomized design to test the differences among the panels. The comparison between means was done employing a Duncan's multiple-range test at 0.05 level of probability. Moreover, the correlation was done between the values of formaldehyde emission resulted from the two methods.

3 RESULTS AND DISCUSSION
3. REZULTATI I RASPRAVA

Table 3 presents the statistical analysis results of both mechanical tests and formaldehyde emission. Average value of MOR ranged from 41 to 51.9 N/mm² (bending parallel to grain) and from 27 to 31.9 N/mm² (bending perpendicular to grain). All the manufactured panels had higher MOR than the requirement level for 15 mm thick panels in accordance with EN 636 (the lower limit value of MOR 23 N/mm²). The range of result of MOE was from 5032 to 5722 N/mm² (bending parallel to grain) and from 2025 to 2536 N/mm² (bending perpendicular to grain). The recommended value of MOE is 1500 N/mm² as described by EN 636. All of the plywood panels had MOE higher than the requirement for EN 636.

Additionally, we monitored changes in the content of formaldehyde for each panel of beech plywood. The values of formaldehyde emission ranged from 0.01 to 0.027 mg/m³ as measured by EN 717-1 method with an average value of 0.016 mg/m³. The results showed that the panel p6 had the highest amount (0.027 mg/m³) while the panel p1 had the lowest amount (0.01 mg/m³) of formaldehyde emission. Similarly to EN 717-1

Table 3 Mechanical properties and formaldehyde emission from beech plywood (15 mm thick) bonded with phenol-formaldehyde resin
Tablica 3. Mehanička svojstva i emisija formaldehida iz bukovih furnirskih ploča debljine 15 mm zalijepljenih fenol-formaldehidnom smolom

Board No.	**MOR,** N/mm²		**MOE,** N/mm²		**Formaldehyde emission** *Emisija formaldehida*	
	A	B	A	B	mg/m³	mg/m²·h
1	47.2±11.4[c] (24)	31.3±9.18[a] (29.3)	5639±993[b] (18)	2536±539[a] (21)	0.01±0.001[e]	0.13±0.008[f]
2	41±6.03[e] (14.7)	31.8±8.08[a] (25.4)	5636±462[b] (8)	2451±462[c] (19)	0.013±0.0008[d]	0.17±0.015[e]
3	41.1±9.82[e] (23.9)	31.9±7.57[a] (23.8)	5032±612[e] (12)	2471±417[b] (17)	0.014±0.0005[cd]	0.21±0.018[d]
4	44.2±5.03[d] (11.4)	30.4±1.75[a] (5.8)	5379±548[d] (10)	2025±65.4[f] (3)	0.015±0.0009[c]	0.25±0.007[c]
5	51.9±5.49[a] (10.6)	25.4±4.17[b] (16.4)	5722±685[a] (12)	2071±2[e] (10)	0.019±0.0017[b]	0.29±0.02[b]
6	49.3±2.8[b] (5.7)	27±7.62[b] (28.2)	5426±385[c] (7)	2360±355[d] (15)	0.027±0.0018[a]	0.33±0.01[a]
mean	45.78	29.63	5472.33	2319	0.016 (0.013 ppm)*	0.23
R^2	0.89	0.70	0.99	0.99	0.96	0.95
P value	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001

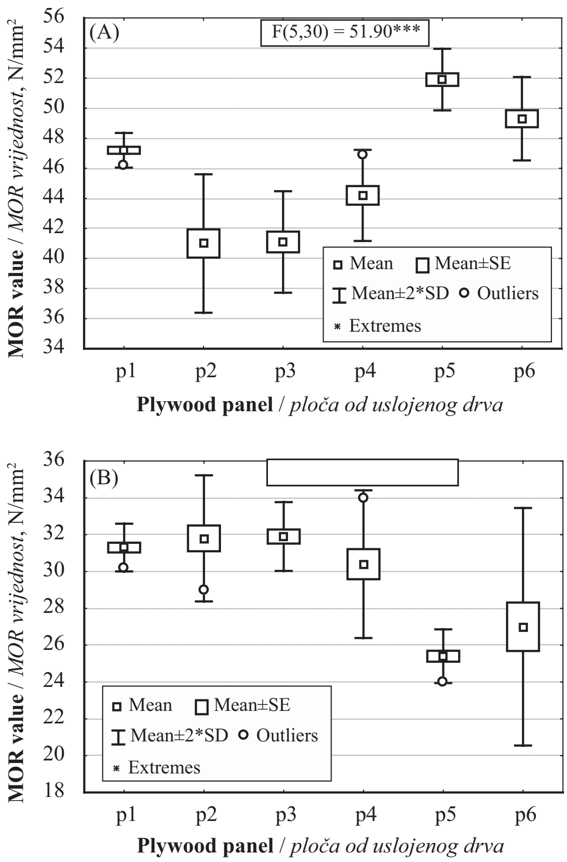
(A): Bending parallel to grain / *savijanje paralelno s vlakancima*; (B): Bending perpendicular to grain / *savijanje okomito na vlakanca*

* At 23 ºC and 1013 hPa, the following relationship exists for formaldehyde measured by EN 717-1: 1 ppm = 1.24 mg/m³ or 1 mg/m³ = 0.81 ppm / *Pri 23 ºC i 1013 hPa za emisiju formaldehida mjerenu metodom EN 717-1 vrijedi odnos: 1 ppm = 1,24 mg/m³ ili 1 mg/m³ = 0,81 ppm.*
Values are mean ± standard deviation (coefficient of variance). / *U tablici su dane srednja vrijednost + standardna devijacija (koeficijent varijacije)*
Means with the same letter within the same column are not significantly different at 0.05 level of probability, according to $LSD_{0.05}$.
Srednje vrijednosti označene istim slovom unutar istog stupca ne razlikuju se signifikantno pri razini vjerojatnosti 0,05.

method, the formaldehyde emission values measured by EN 717-2 had the same trend and ranged between 0.13-0.33 mg/m²·h; however, its values were much higher. This phenomenon could be explained by different conditions used by the two methods.

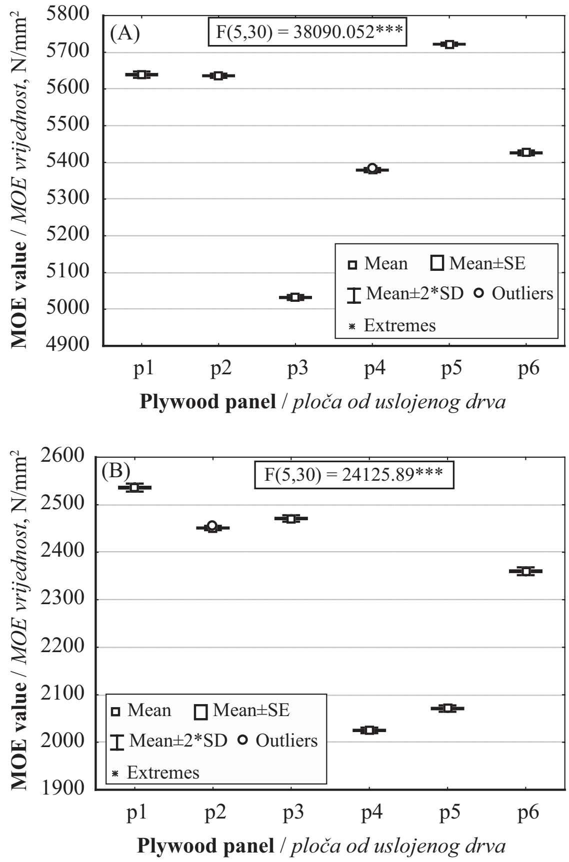
Analysis of variance was used to evaluate the differences between the boards. Statistically it was proven that there was a significant difference (P <0.001) between the means of the boards. The evaluation of the differences among the values of MOR, MOE and formaldehyde emission observed from the six panels of beech plywood is shown in Figs. 1-3. The summary of statistics and comparisons among the means are presented in Table 3 to evaluate the level of significance. The variations among the average of six measurements of MOR for each panel are shown in Fig. 1. The data showed that each of p1 and p2 and p1, p2, p4 and p5 (bending perpendicular to grain) had one value distance (outliers) from the rest of the data. Furthermore, Fig. 2 presents the variations among MOE values. The panel p4 (bending parallel to grain) and p2 (bending perpendicular to grain) had one value as an outlier. The results of formaldehyde emission presented in Fig. 3 show that the panel p6 as measured by EN 717-1 (mg/m³) and the panel p2 as measured by EN 717-2 had only one value outside the rest of the data.

On the other hand, the data presented in Table 3 showed a variety in the relative coefficient of variation (CV %) for the six measurements of each panel for MOR, MOE and formaldehyde emission. For instance, there were variations in the values of CV % for the values of MOR parallel to grain and ranged between 5.7 % (p6) and 24 % (p1) and from 5.8 % (p4) to 29.3 % (p1) for MOR perpendicular to grain. Furthermore, the outliers of the measured MOR, MOE and formaldehyde values for beech plywood showed that there



(A): Bending parallel to grain / *Savijanje paralelno s vlakancima.*
(B): Bending perpendicular to grain / *Savijanje okomito na vlakanca.*

Figure 1 Box-Whiskers plot of MOR value variation among plywood panels (15 mm)
Slika 1. Box-Whiskers dijagram varijacije vrijednosti MOR za furnirske ploče debljine 15 mm



(A): Bending parallel to grain / *Savijanje paralelno s vlakancima.*
(B): Bending perpendicular to grain / *Savijanje okomito na vlakanca.*

Figure 2 Box-Whiskers plot of MOE value variation among plywood panels (15 mm)
Slika 2. Box-Whiskers dijagram varijacije vrijednosti MOE za furnirske ploče debljine 15 mm

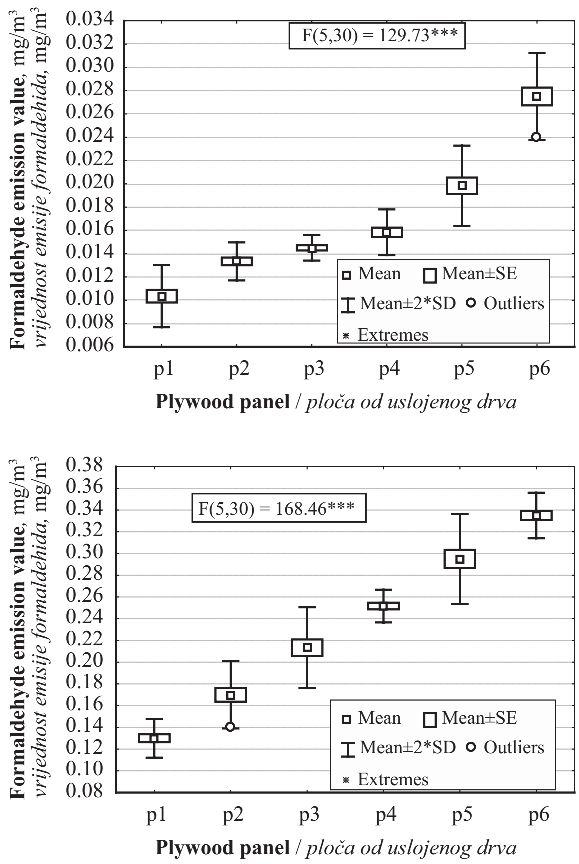


Figure 3 Box-Whiskers plot of formaldehyde concentration variation among plywood panels (15 mm)
Slika 3. Box-Whiskers dijagram varijacije vrijednosti koncentracije formaldehida za furnirske ploče debljine 15 mm

were differences between the values from the six panels for the same kind of wood product, which indicated that some panels had a number of values numerically more distant from the rest of the data than the others.

The data presented measurement points at a distance from the sample mean, i.e. some observations were far from the center of the data. Outlier points can therefore indicate faulty data, erroneous procedures, or areas where a certain theory might not be valid. The inter-panel comparison method used for such analysis depends critically on the quality of the measurements. Under the same standard conditions with the same type of board, significantly higher or lower emission of formaldehyde, MOR and MOE was found in one or more panels than in other boards. Statistical replicability across boards thus becomes an objective yardstick for both the relevance of a behavioral measure and for the estimation of the quality of its measurement.

Even considering that the samples were randomly distributed throughout the plywood panel, which makes it unlikely that the observed variations were due to inter–panel variability, there were some variations in the measured parameters through the tested panel. Consequently, the variations among the values of MOR, MOE and formaldehyde emission were due to sample heterogeneity. For example, the formaldehyde emission values were 0.13, 0.17, 0.21, 0.25, 0.29 and 0.33 mg/m2 h for p1, p2, p3, p4, p5 and p6, respectively. Thus, these results suggested that the most important source of variation is due to heterogeneity of board samples. The obvious results of high, intermediate or low values found among the panels and inter-panel for the same kind of plywood, were in agreements with the previous investigations (Wiglusz *et al*., 2000; Risholm-Sundman *et al*., 2007; Salem *et al*., 2012b; Roffael *et al*., 1979; Bulian *et al*., 2003; Salem *et al*., 2013).

Despite these differences between formaldehyde emission values among the panels, a strongly positive correlation between the formaldehyde emission values measured by EN 717-1 and 717-2 methods was found. The measured values of free formaldehyde from plywood panels with 15 mm thickness had an R^2 value of 0.87 (Fig. 4).

The plywood panels were manufactured with PF resin and formaldehyde emission was close to that of solid wood, because the C-C bonding in the PF resin was very stable against hydrolytic attack (Salem *et al*., 2011b; Böhm *et al*., 2012, Dunky, 2005). Moreover, this low susceptibility to hydrolysis is one of the reasons why PF resins are considered waterproof. At such low levels of free formaldehyde emission, the boards

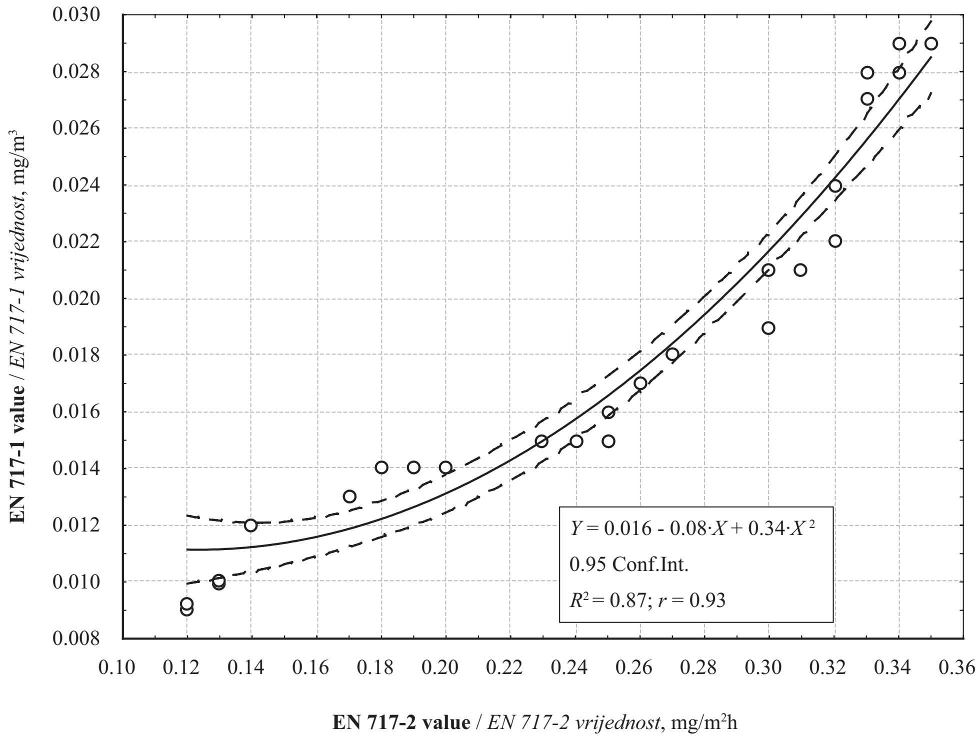


Figure 4 Correlation between the values of formaldehyde emission measured by EN 717-1 and 717-2 methods for beech plywood with 15 mm thickness
Slika 4. Korelacija između vrijednosti emisije formaldehida mjerene metodom EN 717-1 i metodom EN 717-2 za furnirske ploče debljine 15 mm

are considered to be formaldehyde free. It is important to point out here that such low free formaldehyde values may be emitted from the wood itself and the boards are considered formaldehyde free. On the other hand, it was found in the previous studies that the highest amount of formaldehyde released during hot pressing was related to the wood structure (beech wood consists of bigger vessel surfaces) and could be related to the higher density of the beech wood (Salem *et al*., 2012a; Böhm *et al*., 2012).

Simultaneously, the mean values of formaldehyde emission from all the beech plywood panels measured by EN 717-1 were 0.016 mg/m³ (0.013 ppm) and these values are much lower than the requirements of the California Air Resources Board (CARB, 2007) regulations for Phase 1 (≤ 0.08 ppm) and for Phase 2 with the limit value ≤ 0.05 ppm.

Recently, governments of many countries have already imposed or are about to impose regulations limiting the formaldehyde emission from building materials as well as from materials used for the manufacture of furniture, engineered flooring, housing and other industrial products. The emission of formaldehyde in wood products can be minimized during the manufacturing process, or by post treatment and surface treatment of the boards.

4 CONCLUSION
4. ZAKLJUČAK

The results showed significant variations ($P<0.001$) among the beech plywood panels with 15 mm thickness bonded with PF resin, most of them being caused by sample heterogeneity and inconsistency of the values related to inter-panel variation. The values of MOR and MOE were higher than the EN requirements. The concentrations of formaldehyde in these products were below the prescribed limits in the Czech Republic and EU. Moreover, a strongly positive correlation between the formaldehyde emission values measured by EN 717-1 and 717-2 methods was found (R^2 value of 0.87).

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