

## PROPERTIES OF HIGH-DENSITY FIBERBOARDS BONDED WITH UREA-FORMALDEHYDE AND PHENOL-FORMALDEHYDE RESINS

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### ABSTRACT

Urea-formaldehyde (UF) resins are the most common type of resins used in the production of wood-based panels. Despite their numerous advantages, the main drawbacks of these thermosetting aminoplastic resins are the deteriorated water resistance, emission of hazardous volatile organic compounds, and to a lesser extent, the reduced strength. Hence, for producing wood-based panels with increased quality, a modification or partial replacement of UF resins should be performed. There are many previous studies on the improvement of UF resins with melamine-formaldehyde (MF) resins. The partial replacement of UF resins with phenol-formaldehyde (PF) resins has been studied to a lesser extent.

The aim of this research work was to investigate the effects of replacing the UF resin with PF resins on the properties of high-density fiberboards (HDF). The panels were produced at a press factor of 15 s.mm<sup>-1</sup> and a pressing temperature of 220 °C. The resin content in fiberboards was 6%, based on the dry weight of fibres. A complete replacement of UF with PF resin was performed with an increment of 1%. It was found that at 50% content of PF resin in the adhesive system, the panels meet the strictest requirements for load-bearing applications and use in humid conditions. For achieving further improvement of fiberboard properties, the PF resin content should be increased to 83.3%.

**Key words:** wood-based panels; high-density fiberboards; adhesive system; urea-formaldehyde resin; phenol-formaldehyde resin.

### INTRODUCTION

At present, the global production of dry-process fiberboards is significantly higher than the production of wet-process ones. Therefore, high-density fiberboards (HDF) have displaced hardboards (wet-process fiberboards) in terms of production volumes (Antov et al. 2021; FAO).

In the production of dry-process fiberboards, the adhesive bonds have significant importance for the properties of the panels (Hunt et al., 2018). Urea-formaldehyde (UF) resins and, to a much lesser extent, melamine-formaldehyde (MF) resins and phenol-formaldehyde (PF) resins are the most widely used adhesives in this production (Mantanis,

G.I. et al., 2017; Bekhta et al., 2021; Kristak et al., 2021). The dominant use of UF resins is due to their high reactivity, water solubility, good adhesion strength, and significantly lower cost compared to PF resins and MF resins, and due to the lower temperature for polymerisation and significantly faster curing compared to PF resins (Kumar, R.N. and Pizzi, A. 2019a; Kumar, R.N. and Pizzi, A. 2019b; Kumar, R.N. and Pizzi, A. 2019c; Kristak and Réh, 2021). Despite all these advantages of UF resins, they also has some disadvantages, related mostly to the significantly deteriorated dimensional stability and slightly lower mechanical properties of UF-bonded wood-based panels, compared to the panels obtained with MF resins and PF resins

(Kumar, R.N. and Pizzi, A. 2019a). Hence, in case of increased requirements to the properties of the panels, their adhesive system should be modified by adding MF resins or PF resins to adhesive compositions (Pierre-Luis et al., 2006; Pizzi A. et al., 2020). The PF resins are cheaper than MF resins. A disadvantage of PF resins is the need for extended press factor and increased hot-pressing temperature (Pizzi, A. 2003).

The aim of this research work was to investigate the effects of replacing the UF resin with PF resins on the properties of high-density fiberboards (HDF), and establish the optimal ratio of the two resins in the adhesive system, in order to achieve a significant improvement in the properties of the panels with minimal production costs.

#### MATERIALS AND METHODS

In general, the PF resins are used when there are increased requirements to the properties of HDF panels. That is why the total binder content was chosen to be in the upper limit for this type of panel – 6%, based on the dry fibers (Pásztor et al. 2006, Papadopulus, A. N., 2020). The target density of the HDFs was  $920 \text{ kg.m}^{-3}$ , at a target thickness of 4 mm (Pásztor et al. 2006). In order to study the variation of the HDF properties, the panels were produced with a complete replacement of UF resin with PF resin. The substitution was performed with a step of 1%.

Industrially-produced wood fibers were used to fabricate the HDF panels. The pulp was provided by company Welde-Bulgaria AD, Troyan. The wood raw material was composed of two hardwood species, i.e. European beech (*Fagus Silvatica* L.) and Turkish oak (*Quercus Cerris* L.) at the ration of 2:1. Fibers were produced by thermomechanical defibration of wood chips by applying steam treatment at a pressure of 0.8 MPa and temperature of 170 °C. The pulp was stored

in the Laboratory of Press Materials at the Department of Mechanical Technology of Wood at the University of Forestry, Sofia, where it was further dried to 6.2 % moisture content.

The PF resin used was also provided by Welde-Bulgaria AD, Troyan. The PF resin was manufactured by Dynea-Romania and had the following characteristics: dry solids content: 48.0%, viscosity: 364 mPa.s, pH: 6.6, and brix: 72.7.

The UF resin was manufactured and supplied by the factory Kastamonu Bulgaria AD, Gorno Sahrane. The UF resin had a molar ratio of 1.16 and a dry solids content of 64%. This resin was introduced into the pulp at 50% concentration of the adhesive solution. Ammonium sulphate at the content of 2% based on the dry resin was used as a hardener for UF resin. As a water-repellent substance, a wax (paraffin) emulsion was used, at the content of 1% based on the dry fibres.

The introduction of the binders was carried out using a high-speed ( $850 \text{ min}^{-1}$ ) laboratory blender with needle-shaped blades. The adhesive composition was injected through a nozzle with a diameter of 1.5 mm, at a pressure of 0.4 MPa. When the adhesive system was composed of UF and PF resin, the PF resin was sprayed first.

The hot-pressing was performed on a single-opening laboratory press type PMS ST 100, Italy. At present, high-temperature hot-pressing cycles with reduced press factors are used to produce fibreboards (Carvalho, L. and Costa, C. 2003, Anonymous, 2005; Gull et al. 2017; Khayal Os. 2019). Therefore, in this research, the hot-pressing temperature was 220 °C, and the press factor applied was  $15 \text{ s.mm}^{-1}$ . The press factor is consistent with the longer polymerisation time of PF resins (Kumar, R.N. and Pizzi, A. 2019a). The maximum pressure of 4 MPa was reached in 10 s. This pressure was maintained during the first

stage for 5 s. Then the pressure was reduced in 20 s to 0.6 MPa. This pressure value was maintained for 10 s. The press was opened in 15 s. The physical and mechanical properties of the panels were determined according to the European Norms (EN) – EN 310, EN 317,

EN 323. A Zwick / Roell Z010, Ulm, Germany, universal testing machine was used to determine the mechanical properties of the panels.

The manufacturing parameters of the laboratory-produced HDF panels are presented in Table 1.

**Table 1: Manufacturing parameters of HDF panels fabricated from industrial hardwood fibers bonded with UF resin and PF resin.**

HDF Type	Adhesive Type	UF Resin Content, %	PF Resin Content, %
Type A	UF	6	0
Type B	UF + PF	5	1
Type C	UF + PF	4	2
Type D	UF + PF	3	3
Type E	UF + PF	2	4
Type F	UF + PF	1	5
Type G	PF	0	6

**RESULTS AND DISCUSSION**

The results obtained for the density of the laboratory-fabricated HDF panels are presented in Table 2.

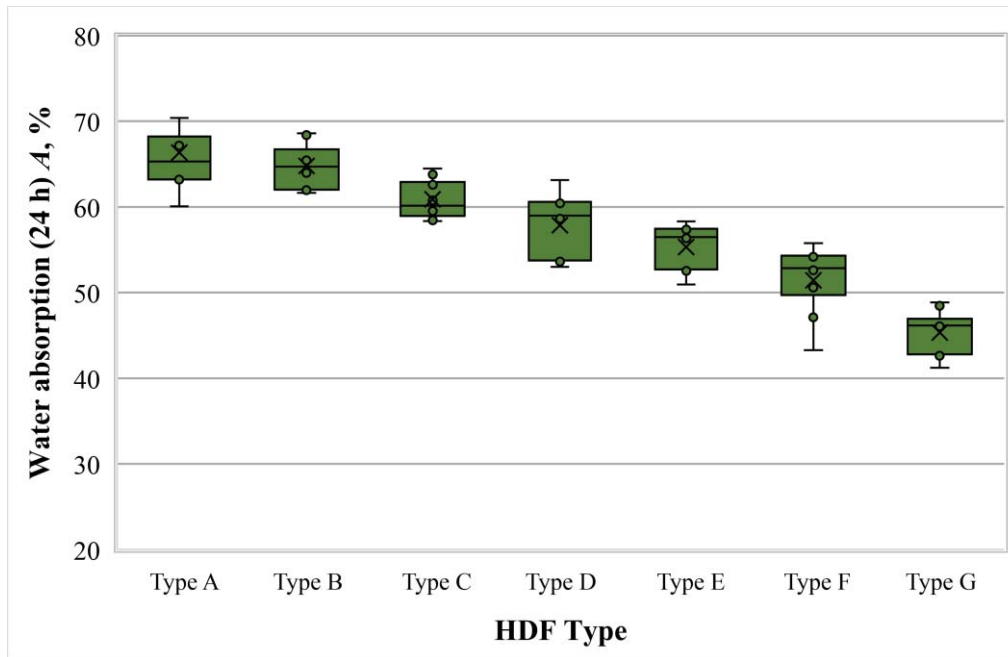
**Table 2: Density of the laboratory panels**

HDF Type	Average (mean value), kg.m <sup>-3</sup>	Standard deviation kg.m <sup>-3</sup>	Standard error, kg.m <sup>-3</sup>	Probability, %
Type A	926	31.41	11.11	1.20
Type B	917	33.58	11.87	1.30
Type C	922	30.35	10.73	1.16
Type D	926	46.33	16.38	1.77
Type E	924	25.43	8.99	0.97
Type F	919	19.20	6.79	0.74
Type G	932	34.84	12.32	1.32

The density of the manufactured panels varied from 919 to 932 kg.m<sup>-3</sup>. Therefore, the variation in this main property is 1.4%, i.e. well below the statistical error. That suggests that the density should not affect the other

physical and mechanical properties of the panels.

A graphical representation of the water absorption (WA) of the laboratory-produced HDF panels is given in Figure 1.



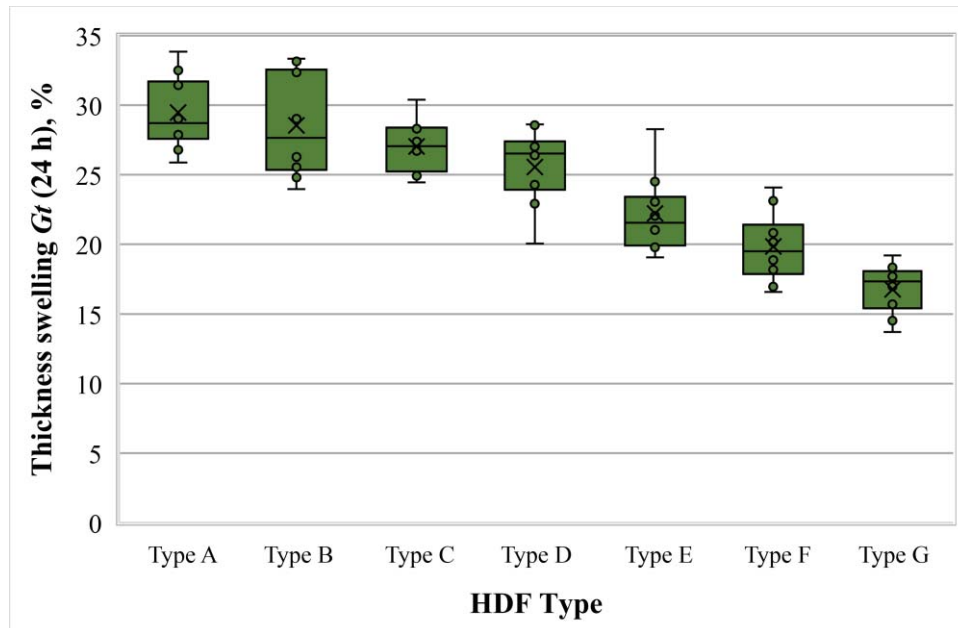
**Figure 1: Water absorption (24h) of the laboratory-produced HDF panels: Type A – 6% UF resin; Type B – 5% UF and 1% PF resin; Type C – 4% UF and 2 % PF resin; Type D – 3% UF and 3% PF resin; Type E – 2% UF and 4% PF resin; Type F – 1% UF and 5 % PF resin; Type G – 6% PF resin**

The complete replacement of UF resin with PF resin resulted in improved WA values of the panels, decreasing from 66.34% to 45.35%. That is, an improvement in this property of HDF panels of 1.46 times was determined. The most significant deterioration in the WA of the panels was observed when replacing 83% and 67% PF resin with UF resin from the adhesive system when the adhesive system is considered 100%. The deterioration of the WA of these HDF panels compared to the previous ones was 1.14 and 1.08 times, respectively. In previous substitutions of UF resin with PF resin, a gradual decrease of WA values was observed by 1.06 times on average. There was also a very small, within the limits of statistical error,

difference between the WA values of the panel bonded with 5% and 1% PF resin and the panels with 6% UF resin, respectively. It can be concluded that in order to achieve a significant improvement in the WA of HDF panels, the PF resin content should be at least 83.3% of the adhesive system and the UF resin content only 16.7%.

The results for the WA of the panels were in accordance with the reported better values of this property obtained when using PF resins compared to those when UF resins are used (Pizzi, A. 2003; Pásztor et al. 2006; Mihailova, J. et al., 2012).

A graphical representation of the thickness swelling (TS) of the laboratory-fabricated HDF panels is shown in Figure 2.



**Figure 2: Thickness swelling (24 h) of the laboratory-produced HDF panels: Type A – 6% UF resin; Type B – 5% UF and 1% PF resin; Type C – 4% UF and 2 % PF resin; Type D – 3% UF and 3% PF resin; Type E – 2% UF and 4% PF resin; Type F – 1% UF and 5 % PF resin; Type G – 6% PF resin**

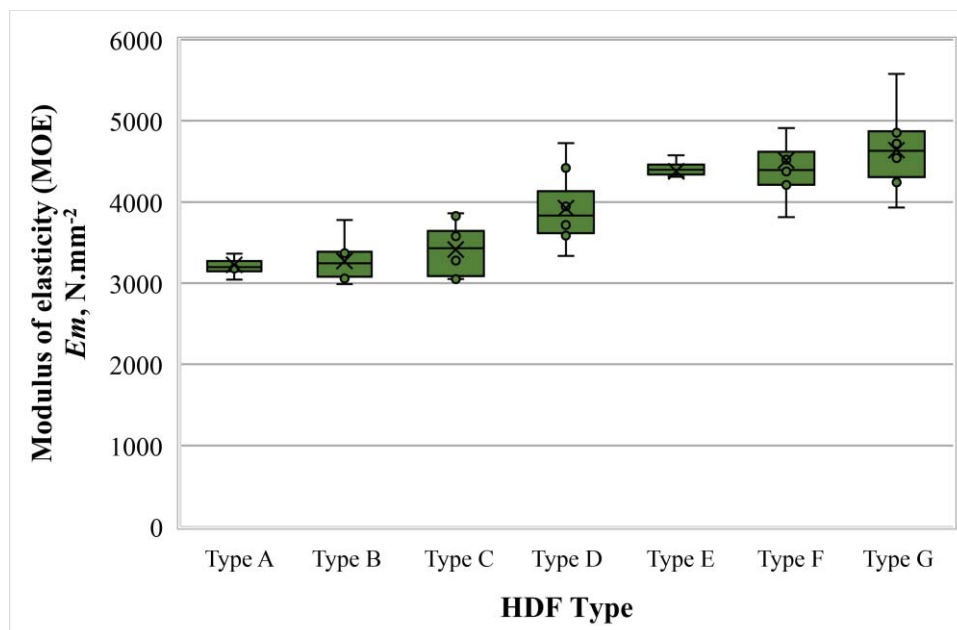
With the complete replacement of UF resin with PF resin, the TS values of the panels were reduced from 29.47% to 16.76%, i.e. a significant improvement of this property by 1.76 times was achieved. The HDF panel, bonded with 5% PF resin and 1% UF resin had 1.18 times higher TS values than the HDF bonded with PF resin only. That is, at only 16.7% of the UF resin and 83.3% PF resin in the adhesive system, a significant deterioration of this property was observed compared to the TS of HDF panels produced with an adhesive system comprising only PF resin. The difference in the property values between the panel with 5% PF resin and 1% UF resin, and 6% UF resin was 1.03 times. That is, again, within the limits of statistical error. A significant decrease of TS values was observed in the adhesive system with more than 50% PF resin. Despite the significantly higher TS values of the HDF panels manufactured with UF resin, all laboratory-fabricated panels met the strictest requirements for the property, namely for panels for load-bearing applications and use in humid conditions – TS values  $\leq 30\%$  (EN 622-5). It

should be emphasised that the HDF with 6% UF resin was on the verge of meeting this requirement but had a TS values significantly below the requirement for panels intended for load-bearing applications and use in dry conditions (TS  $\leq 35\%$ ). These satisfactory TS values were attributed to the relatively high content of binder – 6%, based on the dry fibres.

The obtained results support previous studies on the significantly better TS values of wood-based panels manufactured with PF resins than those manufactured with UF resins (Pizzi, A. 2003; Pásztor et al. 2006; Mihailova, J. et al., 2012).

In terms of mechanical properties, the modulus of elasticity (MOE), bending strength (MOR), and internal bond (IB) strength of the laboratory-fabricated HDF panels were determined.

A graphical representation of the mean MOE values of the HDF panels fabricated with different ratios of UF resin and PF resin in the adhesion system is presented in Figure 3.

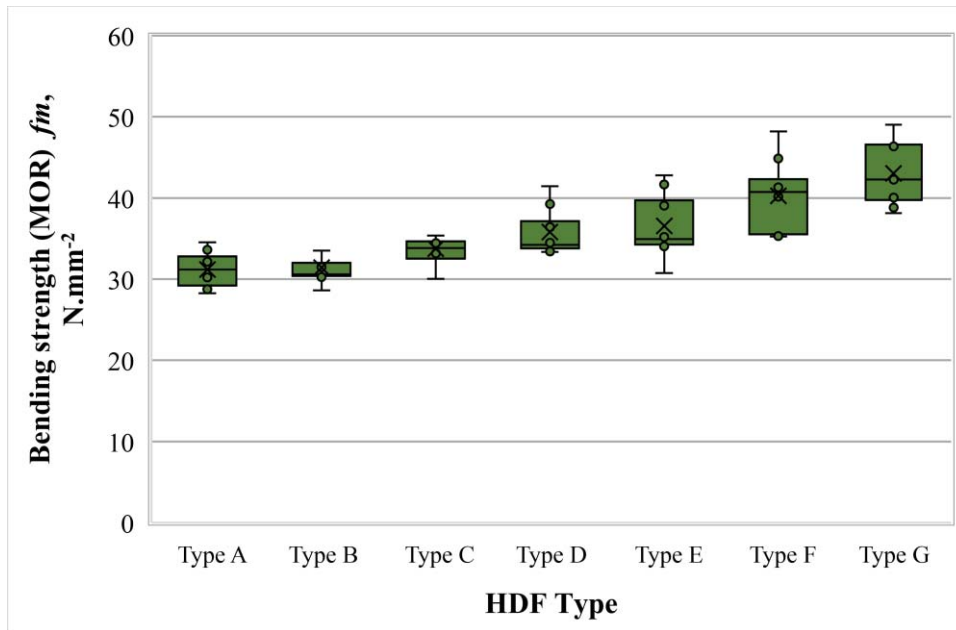


**Figure 3: Modulus of elasticity (MOE) of the laboratory-produced HDF panels: Type A – 6% UF resin; Type B – 5% UF and 1% PF resin; Type C – 4% UF and 2 % PF resin; Type D – 3% UF and 3% PF resin; Type E – 2% UF and 4% PF resin; Type F – 1% UF and 5 % PF resin; Type G – 6% PF resin**

The replacement of UF resin with PF resin in the HDF adhesive system results in increased MOE values ranging from 3223 to 4639 N.mm<sup>-2</sup>, i.e. an improvement of the property by 1.44 times was recorded. The most significant deterioration, by 1.12 times, was observed when the PF resin content was reduced from 4% to 3%. The panel bonded with 4% UF resin and 2% PF resin had 1.15 times lower MOE values compared with the HDF panel fabricated with 3% UF resin and 3% PF resin. In order to achieve a significant improvement of the MOE values of HDF

panels, the PF resin content should be at least 33.7% of the adhesive system. Again, despite the significantly lower MOE values when using UF resin, all manufactured panels met the most stringent requirements for this property – for load-bearing applications and use in humid conditions (MOE  $\geq$  3000 N.mm<sup>-2</sup>) (EN 622-5). That can again be explained by the relatively high resin content.

A graphical representation of the MOR values of the laboratory-produced HDF panels is presented in Figure 4.

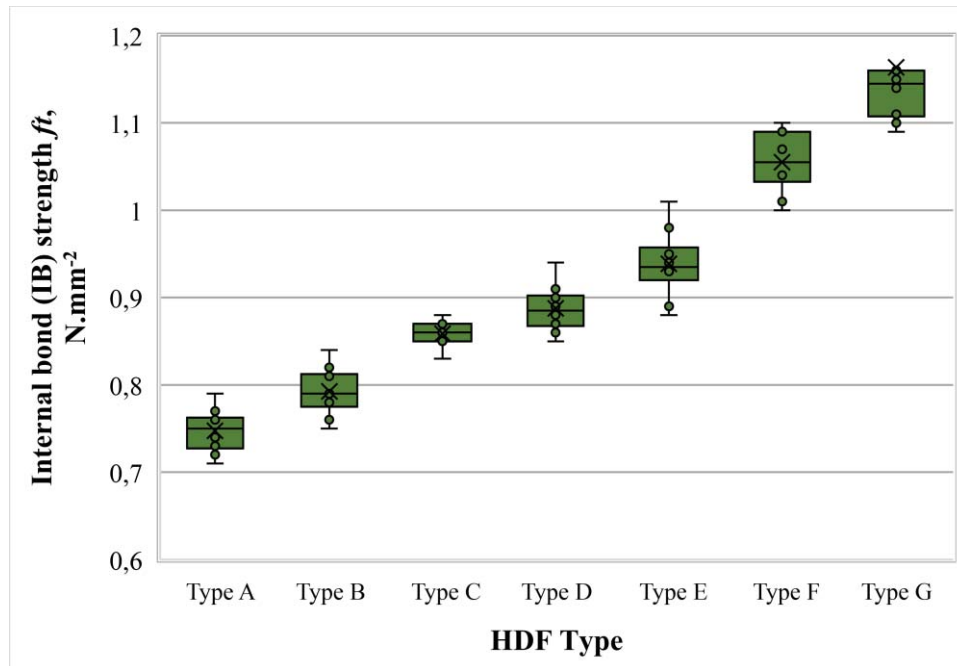


**Figure 4: Bending strength (MOR) of the laboratory-produced HDF panels: Type A – 6% UF resin; Type B – 5% UF and 1% PF resin; Type C – 4% UF and 2 % PF resin; Type D – 3% UF and 3% PF resin; Type E – 2% UF and 4% PF resin; Type F – 1% UF and 5 % PF resin; Type G – 6% PF resin**

With the replacement of UF resin with PF resin in the adhesive system of the panels, the MOR values increased from 31.18 to 43.02 N.mm<sup>-2</sup>. That is, the improvement in this property was 1.38 times. The difference between the panels with 5% UF resin and 1% PF resin, and 6% UF resin was within the statistical error. A gradual improvement of MOR values was observed when replacing UF resin with PF resin up to 33.3% of the adhesive system – the improvement was 1.07 and 1.10 times. The difference between the HDF panel bonded with 4% PF resin and 2% UF resin and the panel produced with 3% PF resin and 3% UF resin was insignificant. Therefore, to achieve a significant improvement in the MOR values of the HDF panels,

PF resin content must be at least 50% of the adhesive system and for even more substantial improvement – at least 83.3%. The panels with more than 50% PF resin content in the adhesive system, i.e. all panels fabricated with PF resin content  $\geq 3\%$ , met the strictest requirements for the property for load-bearing applications and use in humid conditions ( $MOR \geq 34 \text{ N.mm}^{-2}$ ). All HDF panels met the second most stringent requirements for the property – for panels for load-bearing applications and use in dry conditions, i.e.  $MOR \geq 29 \text{ N.mm}^{-2}$  (EN 622-5).

A graphical representation of the mean IB strength of the laboratory-produced HDFpanels is shown in Figure 5.



**Figure 5: Internal bond (IB) strength of the laboratory-produced HDF panels: Type A – 6% UF resin; Type B – 5% UF and 1% PF resin; Type C – 4% UF and 2% PF resin; Type D – 3% UF and 3% PF resin; Type E – 2% UF and 4% PF resin; Type F – 1% UF and 5% PF resin; Type G – 6% PF resin**

The replacement of UF resin with PF resin resulted in increased IB strength values from 0.75 to 1.16 N.mm<sup>-2</sup>, i.e. the overall improvement of that property was by 1.56 times. The most significant improvement of the IB values were determined when the PF resin content was increased above 83.3%. In this case, the increase in IB strength was 1.12 times. The previous improvements in the values had a relatively constant gradient in the range of 1.03 to 1.06 times.

All manufactured panels met the most stringent requirements for this property for load-bearing applications and use in humid conditions ( $IB \geq 0.7$  N.mm<sup>-2</sup>).

The results obtained for the mechanical properties of the HDF panels were comparable with previous studies, which demonstrated better performance of the wood-based panels produced with PF resins than those made with UF resin (Pizzi, A. 2003; Pásztor et al. 2006; Mihailova, J. et al., 2012). This dependence was most clearly seen in the IB strength of the panels.

## CONCLUSIONS

As a result of the study, it was found that the replacement of UF resin with PF resin leads to a significant improvement in all properties of the HDF panels. To achieve a substantial improvement in the properties of the panels, the content of PF resin must be at least 50% of the adhesive composition, and to manufacture panels with very high requirements to its physical and mechanical properties, the PF resin content should be at least 83.3% of the adhesive composition. All panels, manufactured with more than 50% PF resin content, fulfilled the strictest standard requirements to HDF panels intended for use in load-bearing applications and use in dry conditions. In conclusion, the addition of PF resin in the adhesive system for producing HDF panels is justified only if the panels are intended for specialised applications, i.e. load-bearing structures and use in humid conditions. Even in these cases, it is not justified to produce HDF panels only with PF resin. Panels with excellent properties can be

manufactured by partially replacing UF resin with PF resin. In this case, the cost of this type of HDF will be reduced, compared to those using only PF resin, due to the reduced content of the expensive PF resin and the reduction of the press factor.

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### CONFLICTS OF INTEREST

The authors declare no conflict of interest.

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