Using Rape Particles in the Production of Polymer and Lignocellulose Boards

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This study examined the manufacture of flat-pressed wood plastic composites (WPC) using rape chips glued with polymeric diphenylmethane diisocyanate (pMDI). Manufactured boards were of 5-ply type, with 30% polyethylene (PE) content in individual layers. The following properties were assessed: modulus of rupture (MOR); modulus of elasticity (MOE); internal bonding strength (IB); thickness swelling (TS) after 24 hours; water absorption (WA); and noise reduction effectiveness. The tests revealed better physical and mechanical properties of polymer-particle boards than similar polymer and rape boards. It was also demonstrated that a change in mechanical properties of polymer and lignin-cellulose boards is strongly associated with polymer location in individual board layers, and that greater porosity of wood material ensures better polymer anchoring in its structure, when compared with rape straw particles.

Keywords: Rape straw; Polymer; Wood-polymer composites; WPC

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INTRODUCTION

A steadily increasing demand for wood-based materials drives the increasing production volume of these materials. These solutions are usually associated with more efficient use of available raw materials, for example, by production of materials of reduced density, use of recycled materials or of lower quality (Han *et al.* 2006; Hermawan *et al.* 2007; Mirski and Dziurka 2011), or by using other wood species that are currently not used to produce these materials. The replacement of the main raw material with alternate solutions can be described by three fundamental directions: introduction into production species not classified as forest-forming, but common in a given area (Shupe *et al.* 2001; Zaidon *et al.* 2007; Cheng *et al.* 2012), introduction into production of fast-growing tree species and establishing their plantations (Arruda *et al.* 2011; de Araújo *et al.* 2011; Rafighi and Tabarsa 2011; Khanjanzadeh *et al.* 2012; de Almeida *et al.* 2017), introduction into production of species other than trees (Boquillon *et al.* 2004; Guler and Ozen 2004; Li *et al.* 2010; Azizi *et al.* 2011; Li *et al.* 2011; Park *et al.* 2012; Bekhta *et al.* 2013; Dziurka and Mirski 2013).

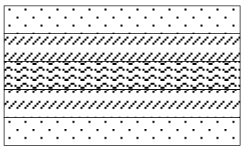
Crop straw, including rape straw, is a material with significant industrial potential. Compared with other types of straw, rape straw is of lower agricultural importance, and it is obtained in a period of early summer, which is conducive to drying. Furthermore, the external surface of stems is coated with a lower amount of waxes, so it is easier to glue with adhesive agents commonly used in the wood-based boards industry. For other straws, problems with gluing are solved by using pMDI (polymeric diphenylmethane diisocyanate) adhesive (Grigoriou 2000; Mo *et al.* 2003; Cheng *et al.* 2013). Previous studies showed that chips created during the cutting of rape straw can substitute for a certain part of pine chips; it is also possible to manufacture boards of standard and of lower density made solely with rape particles (Dziurka and Mirski 2013; Dziurka *et al.* 2005; Mirski *et al.* 2016). Other studies demonstrated that rape particles can be effectively bound with thermoplastic materials (Borusiak and Paukszta 2008; Markiewicz *et al.* 2009). Previously, these materials were manufactured with methods used in the plastics industry, *i.e.*, by molding or extrusion, due to their very high content of thermoplastic materials and small size of filler particles. This method of WPC production ensures uniform heating of a batch. To maintain elastification and forming into a required shape, the material should be cooled after molding. With a small quantity of material in a melting pot, the polymer melts quickly and blends well with the filler. In flat-pressed plastics, this process is more difficult because the heat is transferred into the material first through water vapor and then through layers of the compressed material that has relatively good heat insulation properties. This effect depends both on the type of material and its density. The majority of polymers require plastification temperatures exceeding 100 °C, which can only be reached in a given zone (of a cross-section) of the manufactured plastic after the water evaporates from that zone. Furthermore, the mixture of water vapor and air, which normally migrates into the pressed mat, must be removed to prevent damage to the board during opening of the press. For this reason, use of lignin-cellulose material of low moisture content is preferred. Unlike wood, the majority of additive polymers are hydrophobic compounds (Klason *et al.* 1984; Zaidon *et al.* 2017). Therefore, mechanical adhesion can play a critical role. This process is facilitated by the porous structure of wood, but it may be hindered in other lignin-cellulose materials. The gluing may be improved by using adhesion promoters. These substances usually contain [– $(CO_2)O$–] groups in maleic anhydride, (–Cl) in dichlorotriazine derivatives, or (–N=C=O) in isocyanates. As mentioned above, isocyanates also facilitate the process of board production from particles of annual plants. Considering that compounds of this type are used as binding agents in the traditional technology for production of wood-based composites, and they are also used as adhesion promoters in production of WPC, the authors decided to make use of both these properties to manufacture composites based on rape particles and polymers.

The aim of this study was to examine the manufacture of flat pressed composites (WPC) using rape chips glued with pMDI.

EXPERIMENTAL

Boards were manufactured with particles of baled rape straw received from a farm. The moisture content of the straw was 12.9 ± 0.7%. The straw was shredded in a laboratory mill to obtain particles of a size similar to that of wood chips used as a core layer in type 2 particleboards. The shredded straw was passed through a sorter equipped with two grading sieves, of a mesh of 5 × 5 mm and 0.5 × 0.5 mm. Sieving yielded chips of the following dimensions: length of 14.06 mm (max. 33.37 mm), width of 2.18 mm (max. 3.78 mm), and thickness of 0.62 mm (max. 1.64 mm). Reference boards were manufactured from industrial pine chips also sorted on the grading sieves with mesh of the same size. The mean dimensions of pine chips, excluding thickness, were similar, *i.e.*, length of 13.72 mm (max. 38.43 mm), width of 2.03 mm (max. 3.20 mm), and thickness of 1.04 mm (max. 1.89 mm). The thickness of wood chips was *ca*. 65% greater than that of rape straw particles. The sorted fractions of pine and rape chips were dried to a moisture content of 2% (1.8 ± 0.3%). As the strength of the composite material decreases with an increase in the thermoplastic material share *vs*. the lignin-cellulose material (Boeglin *et al*. 1997; Stark and Berger 1997), the mechanical parameters depend on the quantity of the polymer and its distribution across the board cross-section. For this reason, 5-ply boards were produced with 30% polyethylene (PE) addition in each layer *versus* the dry mass of the lignin-cellulose material. This level of polymer content should ensure increased hydrophobic properties as well as improved mechanical characteristics of the manufactured boards. The board structure is shown in Table 1. The boards were produced as rape (R) and pine (P) versions.

Table 1. Structure of Polymer-enriched Boards

Board structure			PE share per layer			
Layer		Layer share	Board symbol			
			0	**3**	**6**	**9**
			%			
	Surface	14	0	30	30	30
	Intermediate	24	0	0	30	30
	Core	24	0	0	0	30
	Intermediate	24	0	0	30	30
	Surface	14	0	30	30	30

A commercially available polymer was used (Malen E FABS 23-D022, Basell Orlen Polyolefins Sp. z o.o., Płock, Poland), which had an MFR of 1.95 g/10 min at 190 °C/2.16 kg according to EN ISO 1133-2 (2011). The board was 15 mm thick and of density 625 kg/m^3. Three replicate specimens were manufactured for each board type. The pressing conditions were as follows: heat plate temperature, 200 ºC; maximum pressure, 2.5 N/mm^2; and pressing time, 40 s per millimeter of board thickness.

The mat was formed manually on a metal plate. The formed mat was covered with a similar metal plate and inserted into the press. The pressed sheet was removed from the press together with metal plates and placed on a cooling rack to cool (temperature of 70 °C). After conditioning at 20 ± 2 °C and relative air humidity of 60 ± 5%, the following properties were assessed: modulus of rupture (MOR) and modulus of elasticity (MOE) according to EN 310 (1993); internal bonding strength (IB) according to EN 319 (1993); thickness swelling (TS) after 24 h according to EN 317 (1993); and water absorption (WA). The assessments of MOR, MOE, and IB were conducted on 10-12 samples.

As can be seen from the WPC literature review, they can also be used as sound barriers (Santoni *et al.* 2018). Therefore, in this work panels were tested in this respect also. The noise reduction effectiveness of the manufactured boards was evaluated as described by Dukarska *et al.* (2014). The measurements were conducted at the following frequencies from the range specified in the standard EN ISO 10140-1 (2010): 125, 250, 500, 1000, 2000, 2500, and 3150 Hz. A 15 mm-thick MFP board of density of 670 ± 15 kg/m^3 was used as a reference board, as it is manufactured of chips of a structure similar to those used in the study. As advised by Smardzewski *et al.* (2014 and 2015), it was assumed that a few percent difference in board density between MFP and laboratory boards would not significantly affect the noise reduction effectiveness. However, in this case, elastic properties, porosity, and air flow resistivity were of greater importance.

The tests results were analyzed using the dedicated statistical application Statistica 13.1 (StatSoft Inc., Tulsa, OK, USA). The detailed analysis focused on two-factor systems. The normality of the distribution at a level of significance $\alpha = 0.05$ was verified with the Shapiro–Wilk test. The Levene's test was used to evaluate variance equality, while the Tukey's HSD test was used to find significant differences between means.

RESULTS AND DISCUSSION

The values of the modulus of rigidity for rape and pine boards are presented in Fig. 1. The modulus of rigidity of control in polymer-free boards was at the same level of *ca*. 11 N/mm^2. Differences became apparent when some part of the lignin-cellulose material was replaced with polymer. In the pine chip boards, there was a visible increase in the modulus of rigidity. A 30% polymer content in external layers increased this parameter by nearly 45%. This means that the polymer strongly reinforced the board structure in these layers by robustly binding wood particles. Thus, clearly one of the mechanisms for polymer binding to wood chips was activated. When the polymer was introduced into the board structure in deeper layers, the strength increased further; however, the strength of these boards (S6) did not differ significantly from that of boards containing less polymer (S3). When the polymer was introduced into all layers, the previously obtained values decreased, but the strength of such a board was still 27% higher than the control board. This behavior of pine chip boards was consistent with other reports.

The behavior of the boards manufactured of rape straw particles differed noticeably. Their bending strength decreased with the increase in the board polymer content. The static bending strength values obtained for individual variants did not significantly differ from the control board (R0). Therefore, in these conditions of board production, distribution of the thermoplastic material in the board structure does not significantly influence its strength. However, this time the board strength did not increase; thus, a polymer-particle or a polymer-pMDI-particle bond is much weaker than bonds with wood. Similar relations were also observed when the polymer effect on the modulus of elasticity was analyzed (Fig. 2). Although in this case a different or a weaker mechanism of polymer binding to rape particles was even more noticeable. The modulus of elasticity was higher in boards manufactured from rape particles *versus* the boards of pine chips. However, addition of the polymer resulted in its reduction, and for the 30% polymer content in the board mass, the modulus of elasticity in boards manufactured this way decreased to the value of the control pine chips board (S0). This situation possibly results from many factors, apart from the polymer-particle binding mechanism. Rape particles are much thinner than wood chips, so the transfer efficiency of moisture into the board and of gases formed during pressing out of the board is poorer. By replacing the increasing quantity of the material of a very low bulk density with the polymer, the quality of the mat was changed, which was very important and confirmed by results for the tensile strength perpendicular to the plane of the board (Fig. 3). The bulk density of rape particles was lower than the bulk density of pine chips by *ca*. 60% and of the polymer used by *ca*. six times. In boards without polymer or with polymer added only to the external layers, the deeper layers showed a higher resistance during pressing of the mat, and this was reflected in its higher modulus of elasticity. By gradually adding polymer to the deeper layers of the board, their resistance to pressure of the layers on the top was weaker. A similar effect was observed in other studies (Kwon *et al.* 2014). This result was confirmed by the fact that with an increase in the polymer content in the board structure, the tensile strength perpendicular to the plane of the board increased regardless of the type of the lignin-cellulose material used, and the board quality depended on the quality of the core layer in that respect.

In both types of the manufactured board, an increase in the strength was observed, indicating that the polymer also participated in binding raw material particles. The observed changes in board mechanical properties suggested the possibility of manufacturing boards from rape particles additionally bound with polymers.

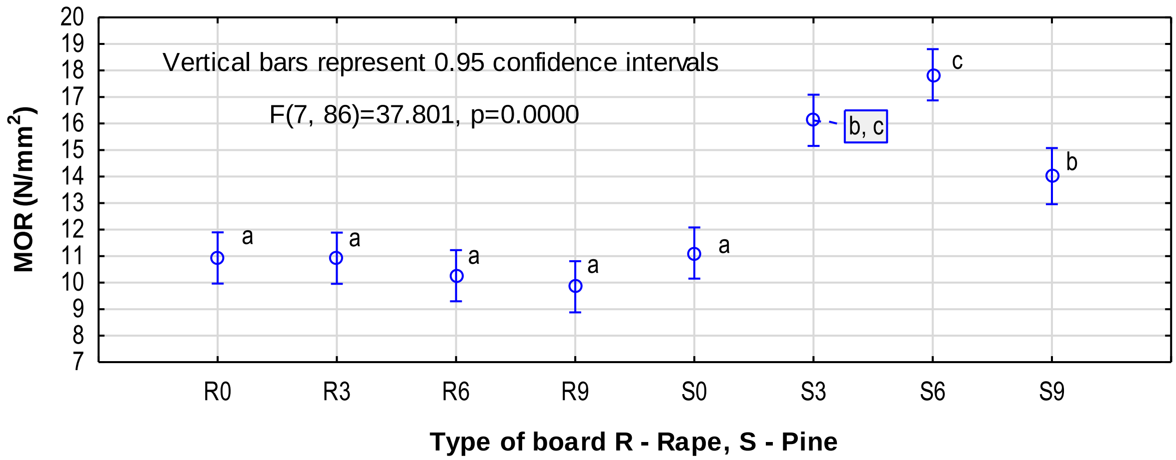


Fig. 1. Modulus of rupture for pine chip boards and for rape straw boards enriched with PE (Letters mark uniform groups determined with the Tukey HSD test)

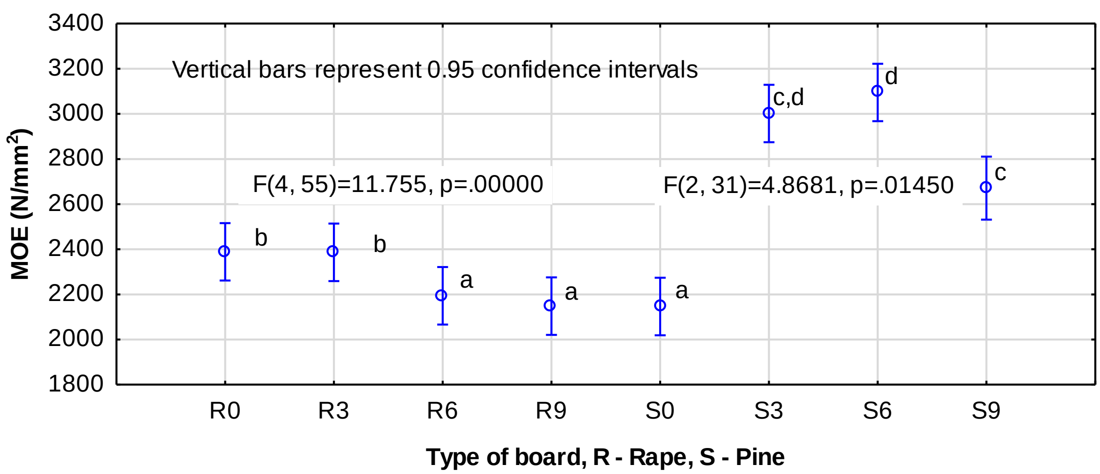


Fig. 2. Modulus of elasticity for pine chip boards and for rape straw boards enriched with PE (Letters mark uniform groups determined with the Tukey HSD test)

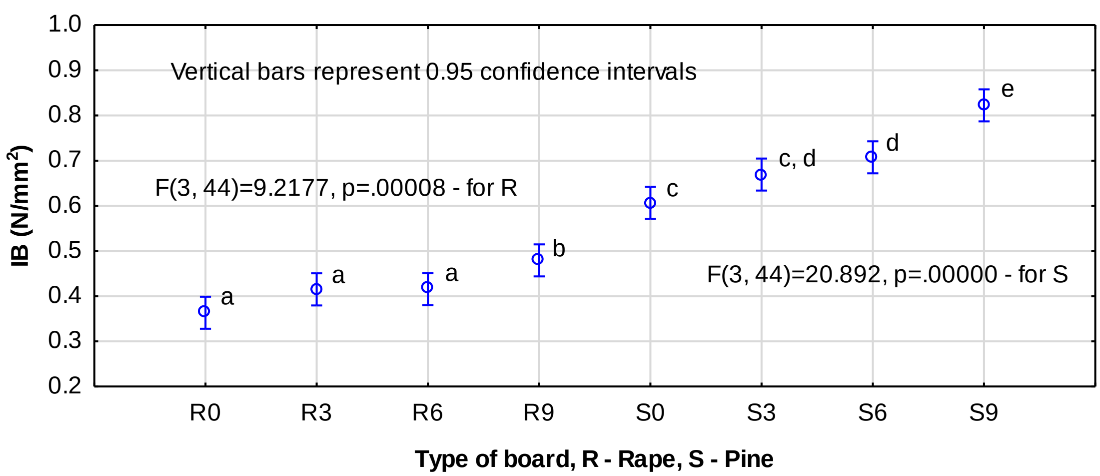


Fig. 3. Internal bonding strength of the board for rape straw boards and for pine chipboards enriched with PE (Letters mark uniform groups determined with the Tukey HSD test)

Notably, the tensile strength perpendicular to the plane of the board was nearly 70% higher in pine chip boards *versus* boards manufactured of rape straw particles. A poorer quality of binding of non-wood particles was previously reported (Ayrilmis *et al.* 2012; Kwon *et al.* 2014; Waelaeh *et al*. 2017). The boards presented in these studies were characterized by increased tensile strength perpendicular to the plane of the board with the increased polymer content, but the strength of these boards was still lower than of the wood-based boards.

The expected positive effects of polymer addition to composites manufactured of lignin-cellulose particles were a reduction in swelling and absorbability. As the two-way analysis shows, the thickness swelling was nearly 70% higher in pine chip boards *versus* boards manufactured of rape straw particles (Fig. 4).

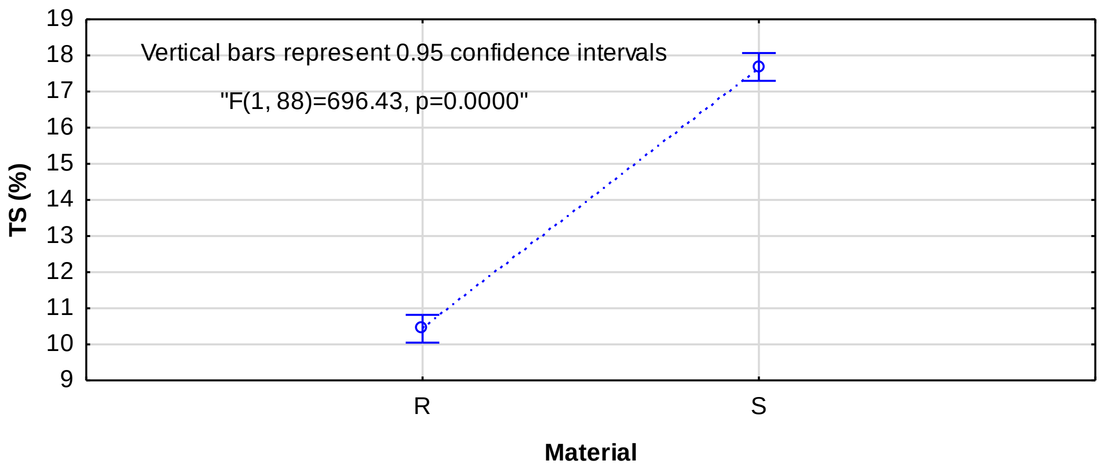


Fig. 4. An effect of the material on board swelling following 24 h immersion in water

The boards manufactured with addition of the polymer were characterized by much lower swelling (Fig. 5).

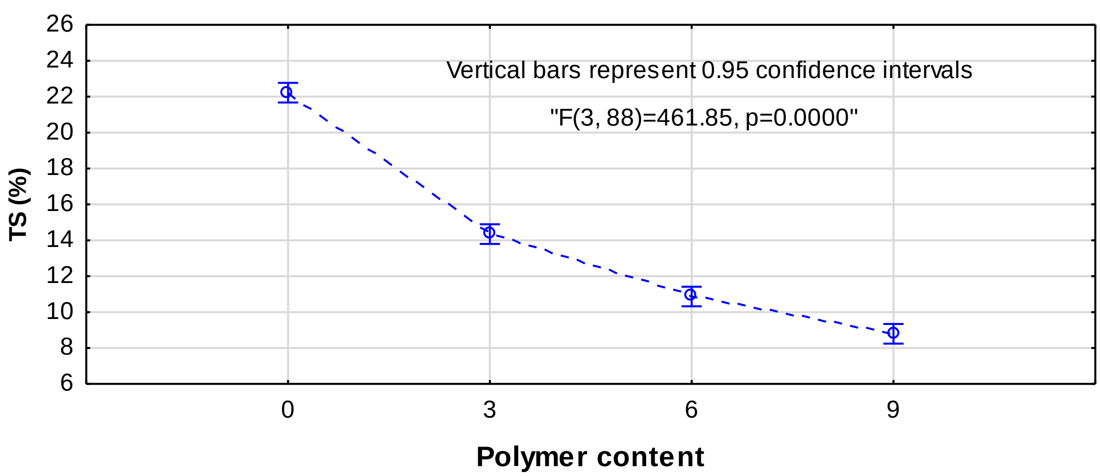


Fig. 5. The effect of the thermoplastic polymer on board swelling following 24 h immersion in water

Regardless of the type of the lignin-cellulose material, when 30% of chips in external layers were substituted with the polymer (*ca.* 10% to the total board mass), the board swelling was reduced by 35%. With the increase in the board polymer content, the swelling of the polymer and lignin-cellulose boards decreased to *ca*. 40% of its initial value when the polymer content approached 30% of the total board mass. This was mainly a result of changes in swelling occurring in boards made of pine chips (Fig. 6).

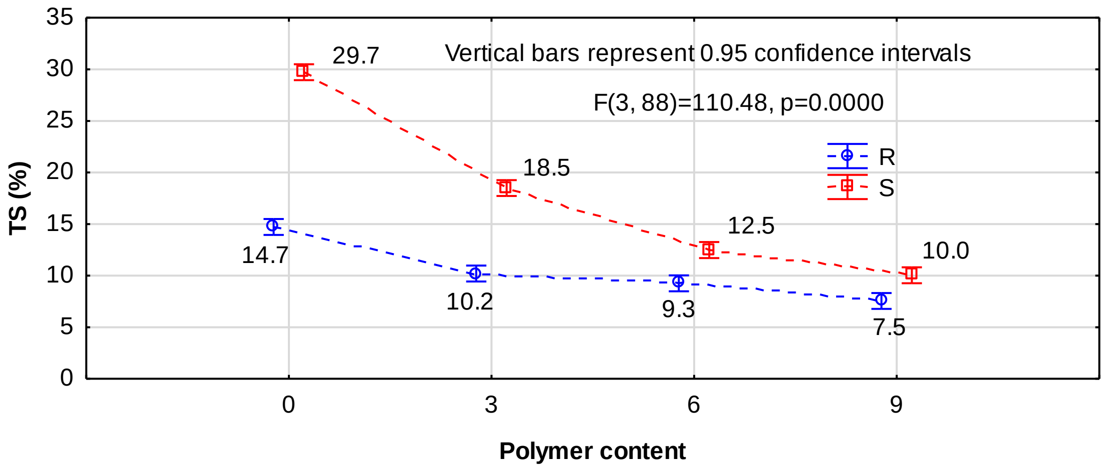


Fig. 6. An evaluation of interactions between the type of board material and the quantity of thermoplastic polymer, and swelling following 24 h immersion in water

In boards of this type, when 30% of chips in external layers were substituted with the polymer, the board swelling was reduced by 38%. When a further 30% of the chip mass was replaced with the polymer, there was an equally pronounced reduction in the board swelling. Although this process slowed with further substitution, these changes were statistically significant (Table 2), and swelling of the board with the 30% polymer content was is nearly three times lower *versus* the control board.

Table 2. A List of Group Uniformity in Terms of Board Swelling Following 24 h Immersion in Water

Uniform groups (HSD Tukey's test, error: Intergroup MS = 1.8120, df = 88.000)								
Board symbol	S0	S3	S6	S9	R0	R3	R6	R9
TS (%)	29.7	18.5	12.5	10.0	14.7	10.2	9.3	7.5
Tukey's HSD	G	F	D	C	E	B	B	A

In boards manufactured with rape particles, changes in swelling were much less pronounced, but in the control board (without polymer) swelling was reduced by half. Furthermore, in rape board with 30% polymer content in its external layers swelling was at the level similar to the pine chip board in which 30% of the total chip mass were replaced with polymer (Table 2). For the similar polymer content in the rape board, the swelling was over two times lower *versus* the reference board (R0) and 25% lower than swelling of the pine chip board. These changes in swelling of boards of both types cannot be explained solely to substitution of the certain percentage of the lignin-cellulose particles with the polymer. For this reason, further studies in this area are needed.

The sound reduction index values obtained for frequencies of 1000 Hz and of 1500 Hz, were similar to each other and independent of the method of board production, raw material type, or board polymer content (Table 3). Certain differences were found at 1250 Hz, where the laboratory boards were characterized by a higher sound reduction index, but its value gradually decreased with the quantity (distribution) of the polymer in the board.

For wave frequencies exceeding 2000 Hz, the laboratory boards, especially those manufactured of rape particles, were characterized by lower values of the sound reduction index. At these frequencies, there was no visible effect of the board polymer content on the value of that index.

Table 3. Acoustic Insulation of Board

Frequency (Hz)	Pfleiderer	Boards manufactured in the laboratory				
	MFP	S0	R0	R3	R6	R9
	Sound reduction index (dB)					
125	13.5	15.8	14.7	14.7	15.9	15.3
250	23.8	22.6	22.6	21.8	21.9	21.6
500	22.2	21.2	22.0	21.9	22.1	22.8
1000	24.8	24.7	23.8	25.6	27.1	26.0
1250	12.7	19.5	27.7	17.4	10.0	9.3
1500	27.3	26.5	27.3	26.8	26.2	26.7
2000	27.1	19.3	13.5	13.7	19.1	16.6
2500	23.0	24.4	20.2	20.9	20.3	21.8
3125	13.1	15.8	9.9	7.9	10.2	7.1

CONCLUSIONS

1. In the assumed manufacturing conditions, the polymer-particle boards were characterized by much better physical and mechanical properties than particleboards. However, in polymer-rape boards, the increase in mechanical properties was only observed for tensile strength perpendicular to the plane of the board. The improvement in the board resistance to water, although very clear, was still lower than in polymer-particle boards.
2. A change in mechanical properties of the polymer and lignin-cellulose boards was strongly correlated with the polymer location in individual layers of the board.
3. The conducted tests, particularly concerning the tensile strength perpendicular to the plane of the board, showed that wood material ensures better polymer anchoring in its structure, when compared to rape straw particles.
4. No clear effect of the polymer location on the noise reduction effectiveness of the boards manufactured from rape particles was found; however, boards containing polymer were characterized by slightly lower noise reduction effectiveness.

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