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EFFECTS OF MANUFACTURE CONDITIONS ON PHYSICAL AND MECHANICAL PROPERTIES OF RAPE-POLYMER BOARDS

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ABSTRACT

The paper presents a method of manufacturing boards composed of lignin-cellulose chips and thermoplastic polymers (waste-based particle polymer composites, WPPC) by means of flat pressing. Rape chips, similar in size to wood chips in traditional particleboards, served as filling material, and polyethylene and polypropylene made up the outer layers. The polymers enriched only the external layers, as this allowed for considerable shortening of pressing time. The resulting boards featured satisfactory properties as compared with control, not polymer covered boards. Our study identified a temperature of 220°C and low moisture content of the polymer-containing layers as favorable for production of this type of boards. We also found out that using a sublayer with higher moisture content not only shortened the pressing time, but also improved the board properties evaluated by a bend test.

KEYWORDS: WPPC, rape straw, mechanical properties.

INTRODUCTION

The resources provided by annual crops are estimated to markedly exceed the demand of wood-based boards industry for the lignin-cellulose materials. Therefore, researchers have tried to make use of this source in the production of particleboards and fiberboards. Potential issues associated with using this type of raw material include its low bulk density and seasonality of supply, which is why considerable storage space needs to be ensured. Extensive research focused on annual plants identified cereal straw as the most promising material, as it has low habitat requirements and may be grown in every climate zone. Moreover, straw provides extensive surface wettability for isocyanate adhesives (Mo et al. 2003, Boquillon et al. 2004). Although isocyanate adhesives are expensive, they easily enhance reactivity and improve properties of UF and

PF resins. The resins may be also added to the adhesive mixture to lower the cost of pMDI (Dziurka and Mirski 2014, Dukarska et al. 2017). Comprehensive research on rape straw (Dukarska et al. 2006, Dziurka and Mirski 2013, Dukarska et al. 2017) showed that wood chips may be partially or completely replaced with rape straw particles glued with a proper type and amount of adhesive.

Waste-based particle polymer composites (WPPC) have been developed for many years. In these materials, wood serves only as a filler and is usually shredded to dust or fine particles. Thermoplastics most often used for WPPC production include polyolefines or PVC. Shaping and bulk dying of WPPCs is easy and they are usually produced by embossing or extrusion. This method of production requires powdered fillers. Replacing wood particles with materials of similar properties is a common trend in this sector of industry. The attempts published so far include such lignin-cellulose particles as rape straw (Paukszta et al. 2014), fibers of sisal, hemp, jute, flax fiber (Kuciel et al. 2010), and rice straw (Sudhakar and Srinivas 2014). Some WPPCs contain lignin-cellulose particles of larger dimensions, i.e. from a few millimeters to a few centimeters. Due to relatively low share of a thermoplastic material, they may be manufactured using a flat pressing method. Flat pressed boards containing larger wood chips and thermoplastic polymers were produced by Pawlicki and Nicewicz (2000), H'ng et al. 2008 and 2010, Ayrilmis et al. 2011 and 2012, Ayrilmis and Jarusombuti 2011, Jastrząb (2010). WPPCs manufactured this way do not require powdered lignin-cellulose materials and they show better physical and mechanical properties, including water-resistance, than standard wood-based materials. They may be also produced using high-throughput processing lines. However, manufacture conditions of such boards differ from the standard ones. The main issues include preferred moisture content of the lignin-cellulose material and pressing time. In general, such WPPC boards contain chips of 1-2% moisture content and their pressing time is about 60 $s \cdot mm^{-1}$ of the board thickness. The literature review and our experience show that the polymer in deeper layers is often not fully converted during shorter pressing times commonly used in the production of wood-based materials. Attempts at shortening the pressing time by enhancing the temperature of the heating plates would cause unfavorable changes in the lignin-cellulose layers directly in contact with the heating plates.

Therefore, the aim of this work was to determine to most favorable conditions for manufacture of flat pressed rape-polymer boards, possibly used instead of wood chip boards, by selecting materials of appropriate moisture content. This is also why we decided to add the polymer only to the external layers.

MATERIAL AND METHODS

The study investigated industrial pine chips and rape straw chips produced in a laboratory. Both types of chips featured similar dimensions. The polymers, i.e. high density polyethylene (HOSTALEN ACP 5231 D) and polypropylene (MOPLEN EP300K) were in the form of 3-5 mm granules.

The boards had three or five layers with the core layer made of pine chips and the external ones from rape straw particles. We produced two types of boards: the first in which the rape layers constituted 50% of a board, and the second comprising 66% of rape in two layers differing in their moisture content. The polymers enriched only the most external layers and they replaced a corresponding weight of rape particles. The polymer share ranged from 0% to 40% with 10% increments. Tab. 1 presents the conditions applied during the board manufacture. pMDI (4% of dry weight of the lignin-cellulose material) was used both as an adhesive and polymer adhesion promoter. We applied the following pressing parameters: pressing time 30 or 20 s·mm^{-1} of the board, board thickness 15 mm, unit pressure 2.5 MPa, temperature of the heating plates 220°C or 200°C, density 580 kg·m^{-3} for three-ply boards and 700 kg·m^{-3} for five-ply boards. Control boards were manufactured solely of rape or pine chips in Y/0/A/2 system. We prepared three variants of the boards: with equal moisture content of rape and pine chips (A), with low moisture content of rape chips (B) and with variable moisture content of rape chips (C).

Each variant comprised four boards, two per series, and the second board was pressed a week later than the first. The WPPC boards were tested based on relevant standards for wood-based boards and the following characteristics were assessed:

- static bending strength (modulus of rigidity - MOR) and modulus of elasticity (MOE) according to EN 310;
- internal bond (IB) according to EN 319;
- water resistance as per V-100 test, according to EN-1087-1;
- swelling after 24 hours according to EN 317.

Tab. 1: The conditions applied during the board manufacture.

Type	Polymer share (%)	Layer share (%)	Layer moisture content (%)	Heating plate temperature (°C)	Pressing time (s·mm^{-1})
Y/0/A/2	0	100	6.5	220	30
X/1/A/2	10	50/50	6.5/6.5	220	30
X/2/A/2	20	50/50	6.5/6.5	220	30
X/3/A/2	30	50/50	6.5/6.5	220	30
X/3/B/2	30	50/50	2/12	220	30
X/3/B/0	30	50/50	2/12	200	30
X/4/A/2	40	50/50	6.5/6.5	220	30
X/3/C/2	30	33/33/33	2/20/8	220	20
X/3/C/0	30	33/33/33	2/20/8	200	20

Y board: Pi (pine) or Ra (rape), X polymer: PP – polypropylene, PE – polyethylene, 0-4 – polymer share 0-40%, A-C – different moisture content of wood and rape chips, 0 and 2 – pressing temperature 200 and 220°C, respectively.

Fig. 1: Samples of manufactured boards (from left to right): PE/3/C/2; Pi/0/A/2; Ra/0/A/2.

The experimental WPPC boards were marked to clearly show that they contain a considerably larger share of lignin-cellulose particles and were manufactured by flat pressing method. Depending on a test, 8 to 15 samples were used and the results were analyzed using Statistica 12.5. We assumed that the boards of density 580 kg·m^{-3} should meet the requirements for P2 boards, and those of density 700 kg·m^{-3} for P5 boards, as set out in EN 312 standard. Photos of manufactured boards are presented on Fig. 1.

RESULTS AND DISCUSSION

Tab. 2 presents mechanical properties of the WPPC boards manufactured from chips of uniform moisture content and subjected to a bend test. The marked properties are consistent with the reports of other researchers (Boeglin et al. 1997, Waelaeh et al. 2017).

Tab. 2: Modulus of rigidity and modulus of elasticity of WPPC boards manufactured from chips of 6.5% moisture content.

Symbol	PP				PE			
	MOR, $N \cdot mm^{-2}$		MOE, $N \cdot mm^{-2}$		MOR, $N \cdot mm^{-2}$		MOE, $N \cdot mm^{-2}$	
	x	υ[1]	x	υ	x	υ	x	υ
Pi/0/A/2	13.9	11.5	2470	7.4	-	-	-	-
Ra/0/A/2	15.1	5.8	2540	5.1	-	-	-	-
X/1/A/2	13.3	10.4	2400	8.3	14.7	8.1	2540	5.1
X/2/A/2	13.0	10.4	2220	5.3	13.3	8.9	2280	6.4
X/3/A/2	12.3	8.9	2030	11	12.4	4.1	1940	9.3
X/4/A/2	11.6	11.2	1870	6.1	11.7	6.1	1900	6.1

[1] Coefficient of variation (%)

Previous studies (Dukarska et al. 2017) showed greater bending strength of rape boards than of pine boards. In this study, the modulus of rigidity and modulus of elasticity of Ra/0/A/2 board were by respectively by 1.2 $N \cdot mm^{-2}$ and 70 $N \cdot mm^{-2}$ higher than those in pine chip board. The difference for the modulus of elasticity was not significant. Polymer enriched boards showed in general lower values of modulus of rigidity and modulus of elasticity than the reference boards Ra/0/A/2 made of rape chips (F(4,50)=26.928, p=0.0000 for MOR for PE; F(4,50)=13.477, p=0.0000 for MOR for PP; F(4,50)=57.567, p=0.0000 for MOE for PE; F(4,50)=44.191, p=0.0000 for MOE for PP), but the differences were significant (Tukey's test) when the polymer share rose above 10%. These values were slightly higher for the boards supplemented with polyethylene but the differences in the modulus of elasticity were insignificant (F(1,80)=4.5085, p=0.03682 for MOR F(1,80)=2.1163, p=0.14965 for MOE). Neither the content nor the type of the polymer significantly affected water resistance measured with V100 test. We found that the values of internal bond were by about 0.06 $N \cdot mm^{-2}$ lower in rape than pine boards. This was probably caused by different density distribution within the board cross section, as indicated e.g. by higher values yielded by the bend test for the rape boards. Also, a drop in strength following a cooking test was similar for both reference boards (Pi/0/A/2 and Ra/0/A/2) and reached about 35%. These differences were probably due to the board formation during pressing, as the bulk density of rape chips is lower than that of pine chips. Therefore, the rape chips show greater compaction in the external layers and are more loosely arranged in the core layer. Pine chips showed a reverse behavior. As the mat is thinner before pressing, the pressing plates more strongly compress deeper layers resulting in a more uniform density of the board cross section.

We saw considerable improvement in water resistance as evaluated by board swelling after 24 h of soaking in water. The changes ranged from 8% to 50% and showed a clear linear trend with coefficient of fit R2>=0.93. A similar improvement in hydrophobicity was also observed by other researchers (Falk et al. 1999, Sellers et al. 2000, Zajchowski et al. 2005). Since the polymer share was between 5% and 20% of the board weight, such a strong reduction in swelling must be due not only to the presence of the hydrophobic component. We therefore assumed that the polymers effectively merged into the structure of rape particles and pMDI was a sufficient adhesion promoter. As a result of the reaction of NCO groups with the OH groups of rapeseeds, the degree of hydrophobization of their surface increases and hence the significant improvement of this property observed (Raj et al. 1988). Tab. 3 presents internal swelling of WPPC boards.

Tab. 3: Internal bond after the boiling test and swelling of WPPC boards made of chips of 6.5% moisture content.

Symbol	PP				PE			
	V100, N·mm^{-2}		TS, %		V100, N·mm^{-2}		TS, %	
	x	υ	x	υ	x	υ	x	υ
Pi/0/A/2	0.17	17	24.2	5.6	-	-	-	-
Ra/0/A/2	0.10	8.4	18.2	5.3	-	-	-	-
X/1/A/2	0.10	11	16.7	8.0	0.11	7.4	15.0	10
X/2/A/2	0.11	13	15.3	15	0.11	9.8	13.9	10
X/3/A/2	0.10	12	14.5	12	0.10	23	10.9	10
X/4/A/2	0.10	14	12.3	17	0.10	13	9.1	14

Reduction in rape chips moisture content from 6.5% to 1.5% and a change of the chip type in the core layer resulted in clear improvement of both mechanical and physical properties of the WPPC boards (Tab. 4). Modulus of elasticity of these boards was similar to that of the control boards (Rp/0/A/2), irrespective of the polymer, and by about 600 N·mm^{-2} higher than MOE in rape chip boards with moisture content of 6.5% and covered with the polymer. Their modulus of rigidity was by about 1 N·mm^{-2} lower than that of the control board but the difference was not significant (F(2, 30)=2.6009, p=0.09085). MOR values were markedly higher than those in the boards from the previous variant (t(1,42)=5.4139, p=0.0000). Only lowering of the pressing temperature by 20°C made the boards produced under these conditions as durable as those discussed above (t(1,42)=-1.3533, p=0.1832). Lowering the pressing temperature did not significantly affect swelling or V100 of the experimental boards. Swelling was still lower than in the control boards containing rape or pine chips alone. V100 test demonstrated a considerable rise in tensile strength perpendicular to the board plane. This was probably due to the presence of pine chips in the core layer, as the values were similar to those of Pi/0/A/2 boards made of pine chips only, and by at least 0.05 N·mm^{-2} higher than in other variants of the three-ply boards (Tab. 3).

Tab. 4: Properties of WPPC boards made of rape chips with moisture content below 2%.

Symbol	MOR, N·mm^{-2}		MOE, N·mm^{-2}		V100, N·mm^{-2}		TS, %	
	x	υ	x	υ	x	υ	x	υ
Ra/0/A/2	15.1	5.8	2540	5.1	0.10	8.4	18.2	5.3
PE/3/B/2	14.0	9.2	2570	8.5	0.19	12	14.0	9.4
PE/3/B/0	12.3	7.6	2550	3.2	0.15	21	16.2	7.7
PP/3/B/2	14.2	9.4	2610	5.0	0.21	6.8	13.1	8.8
PP/3/B/0	13.1	5.8	2550	6.7	0.18	13	14.8	8.5

Weak points of three-ply rape boards covered with polymers include their too low modulus of rigidity and tensile strength perpendicular to the board plane. None of the boards met the requirements specified for the fifth percentile. Despite showing a considerably higher (by ca. 30%) modulus of elasticity and slightly higher modulus of rigidity, the boards containing rape chips of lower moisture content still did not meet those criteria. Mean value of the fifth percentile for those boards was 11.5 N·mm^{-2}, and the highest value for PP/3/B/2 boards was only 12.2. The boards met the requirements of EN 312 standard regarding tensile strength perpendicular to the board plane, and mean value of the fifth percentile was 0.38 N·mm^{-2}. Mean value of the fifth percentile for the WPPC boards made of rape chips alone reached 0.25

A change in WPPC manufacture technology consisting in increasing their mean density and adding layers of higher moisture content caused a clear improvement of mechanical properties determined by a bend test (Tab. 5).

Tab. 5: Properties of five-ply WPPC boards.

Symbol	MOR, N·mm^{-2}		MOE, N·mm^{-2}		IB, N·mm^{-2}			TS, %	
	x	υ	x	υ	x	υ	5.p	x	υ
Ra/0/C/2	20.5b	7.2	3510a	7.5	0.30c	8.9	0.33	19.9a	4.6
PE/3/C/2	22.8a	8.1	3540a	5.0	0.48a	10.5	0.39	17.8c	8.4
PE/3/C/0	20.9b	6.4	3130b, c	3.8	0.44a,b	11.6	0.39	12.2b	8.3
PP/3/C/2	20.4b	8.4	3320a,b	6.6	0.44a,b	11.1	0.37	18.0c	8.9
PP/3/C/0	17.7c	8.1	2950c	7.4	0.39b	8.2	0.34	15.7b	15.9

a, b, c… homogeneous groups according to Tukey's t test ($\alpha = 0.05$), 5.p - value of the 5th percentile.

Mean modulus of rigidity was around 20 N·mm^{-2}, and modulus of elasticity above 3000 N·mm^{-2}. These levels, except for PP/3/C/0 board, exceeded by about 20% the requirements set out in EN 312 standard for P5 boards. However, we did not notice enhanced tensile strength perpendicular to the board plane. Its mean value for four variants of polymer covered boards was similar to that of three-ply boards made of chips with reduced moisture content in the external layer. We added the intermediate layers of greater moisture content to accelerate heat transfer inside the mat. However, the moist chips in external layers probably became more compact and enhanced only modulus of elasticity without affecting compaction of the other layers. We presumably observed two opposite situations. In the second variant, dry external layers pressed on the core layer causing its compaction but impeding heat transfer inside the mat. In the third variant, overheating occurred quickly but it did not cause the core layer compaction and thus the required board strength was not achieved. Gluing degree of 4% was not very high, considering minute size of the particles. Dukarska et al. (2017) obtained satisfactory results for gluing degree of 10%. We could probably limit significant swelling of the boards by using additional hydrophobic agents and increasing the gluing degree.

Statistical analysis showed that WPPC boards covered with PE had more favorable properties than those covered with PP, which was due to higher melting temperature of the polymer's crystalline phase. Irrespective of the polymer type, lowering the pressing temperature by 20°C caused a visible decline in the discussed properties.

CONCLUSIONS

1. Rape straw particles constitute a viable replacement of wood particles in the manufacture of polymer-lignin-cellulose boards.
2. Opting out from using chip-polymer mixture in all layers considerably shortens the pressing time and prevents overheating of the external layers.

3. The study showed that boards with core layer made of pine chips were the most favorable and achieved nearly twice higher internal bond.
4. Moreover, rape chips of low moisture content are preferred for formation of polymer-rape boards, even though they pose a greater technological challenge. The boards with moisture content below 2% in the external layers scored higher in the bend test.
5. Introducing an intermediate layer of higher moisture content enables further shortening of the pressing time, and positively affects modulus of rigidity and modulus of elasticity, probably through a more effective compaction of the external layers.

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