



Some exploitation properties of wood plastic composites based on polypropylene and birch plywood sanding dust

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Abstract. Numerous researchers are closely connected with the production and studies of partially green biocomposites. Various polymer matrices, mainly polyolefines and natural fibres as reinforcement and their combinations, are used. Some scientists use various by-products of the timber industry in preparing wood plastic composites (WPCs). Our work is focused on the investigation of exploitation properties (tensile and flexural strength and modulus, impact strength, microhardness, and water uptake) of the WPCs based on the birch plywood production by-product sanding dust (PSD) and the polypropylene (PP) matrix. Sufficiently good fluidity of the composite melts was noted. To clarify the composites fracture mechanism SEM studies were used. For modifying the WPCs maleated polypropylene (MAPP) was used as the coupling agent. It was shown that the additions of PSD and MAPP had a positive influence on the exploitation properties of WPCs based on a PP.

Key words: composites, polypropylene, birch plywood sanding dust, modification, exploitation properties.

INTRODUCTION

During the last 20–30 years, many researchers have studied exploitation properties of wood polyolefine composites (WPCs) [1–28]. The most useful among the wide selection of polyolefines is polypropylene (PP) [2–7, 9–28], but as a reinforcement of WPCs various wood waste materials comprising lignocellulose fibres are often used [2–28]. Such materials also include wastes that arise for example in plywood production [5–8, 13, 26]. Numerous researchers have tried to clear up the influence of wood fibres on different exploitation properties such as physical–mechanical parameters [1, 2, 8, 11, 12, 14], water resistance [19, 28], and technological properties [27] of WPCs. All these properties

considerably depend on the type of fibres, the length and l/d ratio [18, 24, 25], and the interaction mechanism on the surface wood fibre polypropylene matrix [3, 16, 17, 18, 21, 25].

Some authors [13, 21, 26] have studied the positive influence of wood saw dust on properties of PP composites and have tried to utilize the recycled PP matrix [19, 22, 23, 28] for preparing WPCs. Our earlier studies [4–7] showed that the plywood production by-product sanding dust (PSD) is a promising reinforcement of the PP matrix. The additions of up to 40 wt% PSD increase the tensile and the flexural modulus, but decrease the deformation ability of the PP matrix, the impact strength, the water resistance, and the fluidity of the composite melts. Modification of the composites with the coupling agent maleated polypropylene (MAPP) (up to 5–7 wt%) significantly improves all the above-

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mentioned properties: the tensile and the flexural strength increase 1.7–2.2 times and the tensile and the flexural modulus up to 3 times compared with the unmodified PP + 40 wt% PSD composite. The impact strength values increase from 8.8 kJ/m² up to 13.8–14.4 kJ/m². The WPCs with 40 wt% PSD practically do not swell in the water and the total amount of the absorbed water after 600 h water uptake is no more than 6%, but in the presence of 5 wt% MAPP decreases to 2.2%. Also a significant increase of the microhardness (2 times) of the samples was observed for the composite with 40 wt% PSD. This means that it is possible to use these systems for the production of WPC materials and different products from WPCs.

In this work we approbate PSD for reinforcing PP. This by-product contains wood fibre fractions of different length: longer than 500 μ (1.04%), 250–500 μ (32.16%), and shorter than 250 μ (66.8%) [8]. The goal of this study was to determine the influence of this new type of by-product (from Latvijas Finieris JSC) on the composites based on a virgin PP. The physical–mechanical properties (tensile and flexural strength and modulus, impact strength, and microhardness) and other exploitation properties such as water resistance as well as the fracture mechanism of the composites were studied.

MATERIALS AND METHODS

As the polymer matrix virgin PP type Mosten MA-712 (MF = 12 g/10 min) was used. The content of the filler (PSD) in the composites was 30, 40, and 50 wt%. As an interfacial modifier Licocene PP MA-7452 (1, 3, and 5 wt%) granules were utilized. The composites were prepared by mixing in a two-rolls mill (temperature 180 °C, time 12 min), then cooled at room temperature, granulated in a knife granulator, and pressed into 1 mm thick sheets (pressure 5 MPa, time 3 min, $T = 180$ °C) for the tensile tests (EN ISO 527) and the microhardness measurements by Vickers M-41 at the load of 200 g. The flexural and the impact strength tests (EN ISO 178 and ASTM D 256 M, respectively) were done for standard specimens; bars were produced by injection moulding

($T = 180$ °C, time 1.5 min, $P = 5$ MPa). The fluidity of the melt (melt flow index (MFI) at 230 °C, 5 kg) was estimated by the standard ASTM D 1238. The water resistance measurements were made according to the standard ASTM D 570-88 at +23 °C. Scanning electron microscope (SEM) studies were carried out and the fractured surface of specimens was examined with a TESCAN TS 5136 MM and VEGA TC computer software. Sputter coater EMITECH K 550X for applying an Au layer on the analysed surface was used. SEM pictures were taken at 200, 500, and 2000 \times magnifications.

RESULTS AND DISCUSSION

The earlier studies [4–7] of the influence of different types of plywood production by-products on physical and mechanical properties of unmodified virgin PP showed that the investigated composites deformation ability decreases with an increase of the filler content, but the tensile and the flexural modulus increase with an increase of the by-product content in PP. The flexural modulus increases from 1500 MPa for a virgin PP up to 2600–3400 MPa for the filled systems. In addition, all composites maintain a good enough fluidity: the MFI value changes from 4.6 g/10 min for a virgin PP to 0.6–1.11 g/10 min for filled systems (filler content 40–50 wt%) [7], which is sufficient to process these materials with the traditional polymer processing methods. For example, for the extrusion process the lowest permissible values of the MFI are about 0.1–0.3 g/10 min, but for compression moulding it could be lower than 0.1 g/10 min. Possibilities of processing the high-filled WPCs by traditional polymer processing methods are shown in [25,26].

The presented results of the studies of the physical–mechanical properties of PP with different contents of the new type of PSD are listed in Table 1. From the completed measurements we can conclude that in general PSD has a positive influence on the PP matrix properties. The flexural strength changes very little, but the flexural modulus increases up to 2.42 times and the microhardness

Table 1. Physical–mechanical properties of the composites based on polypropylene (PP) and birch plywood sanding dust (PSD)

PSD, wt%	MFI, g/10 min	Tensile			Flexural			Impact strength A , kJ/m ²	MH, MPa
		σ_t , MPa	E_t , MPa	ϵ_t , %	σ_f , MPa	E_f , MPa	ϵ_f , %		
0	12.0	32.4	469.0	864.5	51.45	1960	5.67	10.73	107.5
30	7.65	18.3	434.6	7.42	50.83	3633	3.02	10.54	117.3
40	6.54	15.6	446.5	5.45	50.36	3978	2.31	7.67	141.5
50	5.41	10.9	510.7	5.09	49.70	4740	1.80	6.91	139.4

1.3 times. The tensile strength decreases considerably (3 times) and the impact strength decreases by about 36%. The MFI values also diminish till 5.41 g/10min for the system with 50 wt% PSD. Nevertheless, we have to take into consideration that the polarity of wood fibres and the PP matrix differ significantly. The utilized type of the PSD contains a larger amount of fine particles compared to the PSD used in our previous studies [4–7]. Due to this fact the fibre of the new by-product has also a higher specific surface. Therefore, we can expect as in [17,18,24] that the interaction on the interfacial surface will be rather weak and as a result, the structure of the prepared WPCs could have a high heterogeneity and some properties need not be so good. Bearing in mind these expectations and our previous investigation results [4–7], 40 wt% of the PSD in PP was chosen as an optimal content.

One of the best methods for intensifying adhesion processes between the wood fibres and the PP matrix is to use coupling agents such as maleated polypropylene (MAPP) [2,16,21,26]. Due to the addition of MAPP, a common tendency of an increase of the mechanical and other exploitation properties of the PP + 40 wt% PSD composites can be observed (see Table 2). The addition of 3–5 wt% of MAPP to PP + 40 wt% PSD system improves practically all exploitation properties of these composites. The tensile and the flexural strength increase up to 27.4–28.0 MPa and 53.18–53.56 MPa, respectively (with 3 and 5 wt% MAPP). Obviously, the content of 3 wt% of MAPP is sufficient to improve the interaction on the surface between the polar wood fibre and the nonpolar PP matrix while the rest of the MAPP has not reacted with the polar –OH groups of the cellulose that contains wood fibre. This free, uncombined MAPP can work also as a plasticizer and decrease mechanical properties of the WPCs. The decrease of the tensile and the flexural modulus by about 30–35% suggests this possibility. The impact strength, on the contrary, significantly increases: from 10.73 kJ/m² for unmodified material up to 13.96 kJ/m² and 14.48 kJ/m² for modified systems. This phenomenon indicates that in the presence of MAPP WPCs have become more elastic

in comparison with the unmodified material. Therefore, the optimal concentration range of MAPP could be 3–5 wt%. At the same time, these composite melts maintain also rather good fluidity (MFI values are 5.61 and 5.23 g/10 min). The greatest microhardness is shown by the composites containing 40 wt% of PSD (141.5 MPa) and 50 wt% of PSD + 5 wt% MAPP (145.4 MPa). Some of our results correspond to results obtained by other authors [10–16].

The addition of the interfacial modifier (3–5 wt% MAPP) also promoted an increase of the water resistance (see Fig. 1). The absorbed amount of the water after 1000 h of water uptake was only 2.6–2.7% (Fig. 1, curves 3 and 4) in comparison with the unmodified system (curve 1), which absorbed about 4.5%. Similar results are reported in [5,19,28]. It is necessary to note that the water sorption kinetics of the systems with 3 and 5 wt% of MAPP practically coincided (curves 3 and 4). This means that the 3 wt% concentration of MAPP is sufficient to provide a good water resistance of the investigated WPCs. Bearing in mind the cited studies of the exploitation properties of the modified WPCs and the prices of MAPP we recommend 3 wt% as an optimal content of MAPP in these composites.

We also tried to clarify the fracture mechanism of the investigated WPCs with SEM studies. The results of the studies of the fractured surfaces of the impact strength of the tested samples are presented in Fig. 2. The SEM pictures indicate that the coupling agent MAPP has a positive influence on the strengthening effect of the PP matrix. The SEM picture of the PSD (Fig. 2A) shows that the particles of the wood fibres are of irregular shape and different sizes. Most of the particles are smaller than 250 μ and the surface of the fibres is rough with deep hollows. In the SEM images replicas of a fracture surface of an unmodified material (Fig. 2B, C) and partially or completely uncovered wood particles can be observed. In addition, the polymer matrix is significantly deformed. The length of the whiskers of PP is about 100 μ (Fig. 2C), which indicates a high deformation ability of the PP and a poor interfacial interaction between the wood fibres and the PP matrix.

Table 2. Physical–mechanical properties of the composites based on polypropylene + 40 wt% birch plywood sanding dust (PSD) modified with maleated polypropylene (MAPP)

MAPP, wt%	MFI, g/10 min	Tensile			Flexural			Impact strength <i>A</i> , kJ/m ²	MH, MPa
		σ_t , MPa	E_t , MPa	ϵ_t , %	σ_f , MPa	E_f , MPa	ϵ_f , %		
0	6.54	15.6	446.5	5.45	50.36	3978	2.31	10.73	141.5
1	5.46	21.5	342.8	4.13	49.87	2320	3.21	9.99	143.3
3	5.61	27.4	317.2	6.19	53.56	2378	3.94	13.96	144.2
5	5.23	28.0	303.3	7.23	53.18	2525	4.52	14.48	145.4

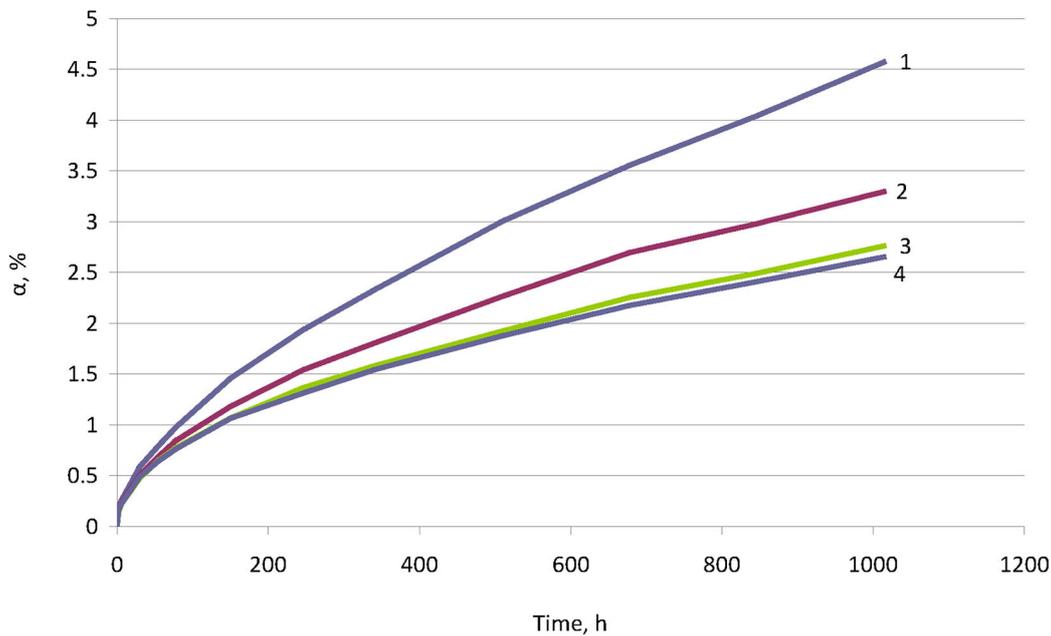


Fig. 1. Water absorption kinetics of the polypropylene (PP) composites: 1 – PP + 40 wt% PSD, 2 – PP + 40 wt% PSD + 1 wt% MAPP, 3 – PP + 40 wt% PSD + 3 wt% MAPP, 4 – PP + 40 wt% PSD + 5 wt% MAPP. PSD – birch plywood sanding dust, MAPP – maleated PP.

As expected, the additives of the coupling agent MAPP strengthened the border surface between the PP matrix and the wood fibres (Fig. 2D, E). This provides evidence that the border surface of the interface is only partially broken, but the fine particles in the polymer matrix hold very well. Furthermore, in the presence of MAPP the polymer matrix is less deformed. The length of the whiskers considerably decreases for the composites with 1 wt% of MAPP (Fig. 2D) and particularly with 3 wt% of MAPP (Fig. 2E). This evidently points to a decrease of the deformation ability of the PP matrix due to the strengthening action of MAPP.

CONCLUSIONS

It was shown that the birch plywood production by-product sanding dust (PSD) can be successfully used without additional fabricating in the production of wood polyolefine composites (WPCs). Due to their improved properties, such materials could be competitive with the currently produced WPCs. The optimal concentration of PSD in polypropylene (PP) could be 40 wt%. The PSD additives in the PP composites increase their flexural modulus 2.4 times and microhardness 1.3 times. The modifying of the unmodified PP composite (PSD

content 40 wt%) with maleated polypropylene waxes (MAPP, with the optimal content of 3 wt%) improved its physical–mechanical properties (tensile, flexural, and impact strength and microhardness). The changes of the rheological properties (melt flow index values) were negligible. The amount of absorbed water of the samples decreased almost two times. This fact confirms that adhesion interaction between the wood fibres and the PP matrix in the modified material had improved. The SEM studies also confirmed previous suggestions and display a positive influence of MAPP on the strengthening of the investigated WPCs due to the improving of some exploitation properties.

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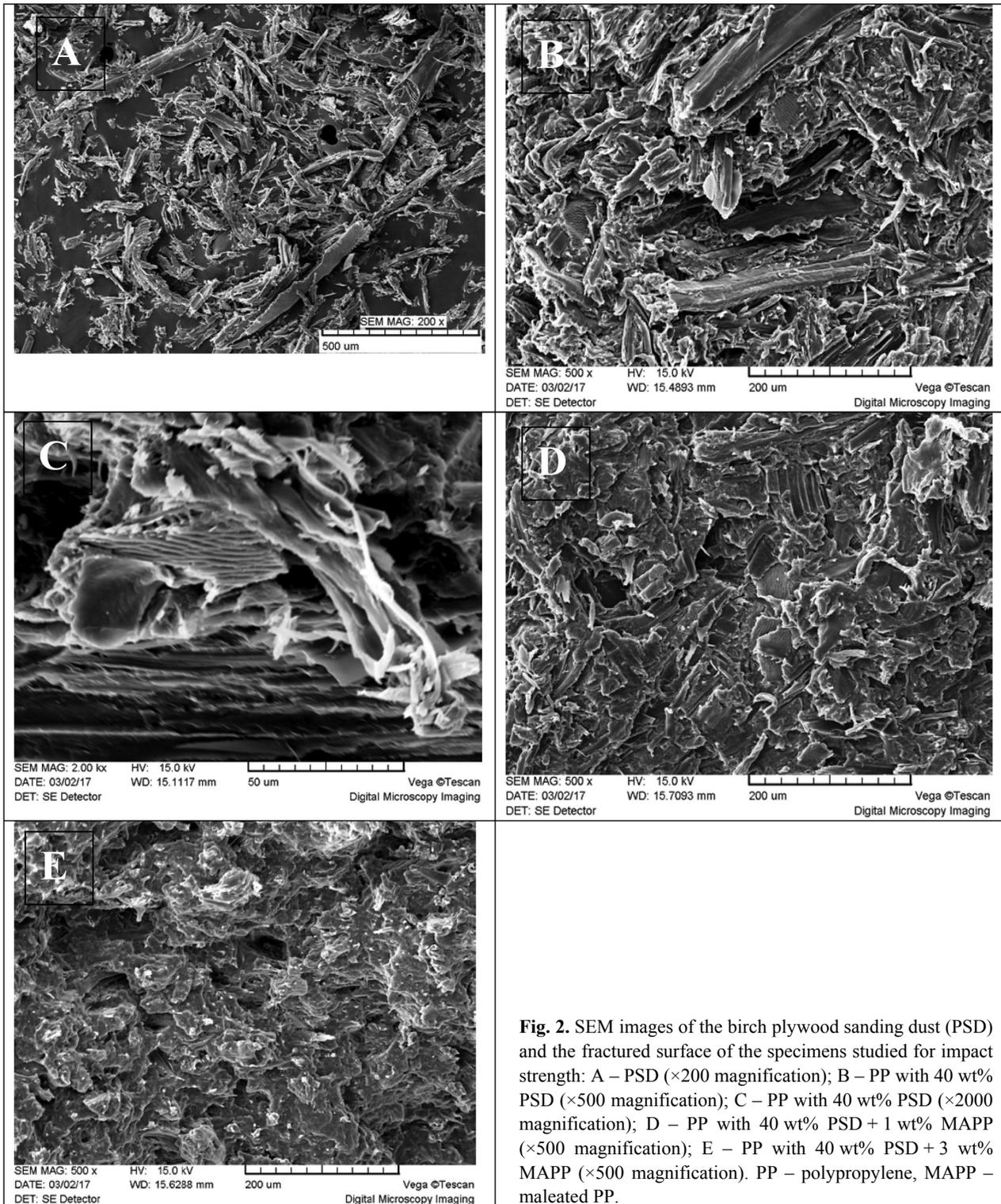


Fig. 2. SEM images of the birch plywood sanding dust (PSD) and the fractured surface of the specimens studied for impact strength: A – PSD ($\times 200$ magnification); B – PP with 40 wt% PSD ($\times 500$ magnification); C – PP with 40 wt% PSD ($\times 2000$ magnification); D – PP with 40 wt% PSD + 1 wt% MAPP ($\times 500$ magnification); E – PP with 40 wt% PSD + 3 wt% MAPP ($\times 500$ magnification). PP – polypropylene, MAPP – maleated PP.

REFERENCES

- Raj, R., Kokta, B., Maldas, D., and Daneault, C. Use of wood fibers in thermoplastics. VII. The effect of the coupling agents in polyethylene–wood fibre composites. *J. Appl. Polymer Sci.*, 1989, 7, 1089–1103.
- Kokta, B., Raj, R., and Daneault, C. Use of wood flour as filler in polypropylene: studies on mechanical properties. *Polym.-Plast. Technol.*, 1989, 28(3), 247–259.
- Maldas, D. and Kokta, B. Role of coupling agents on performance of wood flour-filled polypropylene composites. *Int. J. Polym. Mater.*, 1994, 27(1–2), 77–88.
- Nwabunma, D. and Thein Kun, T. (eds). *Polyolefin Composites*. 3M Company, Wiley-Interscience A. John Wiley and Sons INC Publications, 2007.
- Kajaks, J., Kalnins, K., Uzulis, S., and Matvejs, J. Physical and mechanical properties of composites based on polypropylene and timber industry waste. *CEJE*, 2014, 4(4), 385–390.
- Kajaks, J., Kalnins, K., Uzulis, S., and Matvejs, J. Some exploitation properties of wood plastic hybrid composites (WPHC) based on polypropylene and plywood production waste. *Open Engineering*, 2015, 5(1), 457–464.
- Kajaks, J., Kalnins, K., Uzulis, S., and Matvejs, J. Some exploitation properties of wood plastic composites based on polypropylene and plywood production waste. *Key Eng. Mater.*, 2017, 721, 48–52.
- Kajaks, J., Kalnins, K., and Naburgs, R. Wood plastic composites (WPCs) based on high density polyethylene and birch wood plywood production residues. *Int. Wood Prod. J.*, 2018, 9(1), 15–21.
- Ramakrishna, M., Kurmar, V., and Singh, Y. N. Recent development in natural fibre reinforced polypropylene composites. *J. Reinf. Plast. Comp.*, 2009, 28, 1169–1189.
- Sobczak, L., Lang, R. W., and Haider, A. Polypropylene composites with natural fibers and wood – general mechanical properties. *Compos. Sci. Technol.*, 2012, 72(5), 550–557.
- Bulylina, S., Mactikka, O., and Karki, T. Properties of wood fibre–polypropylene composites: effect of wood fibre source. *Appl. Compos. Mater.*, 2011, 18(2), 101–111.
- Ashori, A. Study of mechanical properties of wood fiber–polypropylene composites. *Adv. Mater. Res.*, 2010, 123–125, 1195–1198.
- Lou, C.-W., Lin, C.-W., Huang, C.-H., and Li, T.-T. Preliminary study of polypropylene/sawdust green composites. *Adv. Mater. Res.*, 2012, 557–559, 334–337.
- Perez, E., Fama, L., and Pardo, S. Tensile and fracture behaviour of PP/wood flour composites. *Compos. Part B-Eng.*, 2012, 43, 2795–2800.
- Kumar, V., Tyagi, L., and Sinha, S. Wood flour-reinforced plastic composites: a review. *Rev. Chem. Eng.*, 2011, 27(5–6), 252–264.
- Mendez, I. A., Vilaseca, F., and Pelach, M. A. Evaluation of reinforcing effects of ground wood pulp in the preparation of polypropylene based composites coupled with maleated polypropylene. *J. Appl. Polymer Sci.*, 2007, 105, 3588–3596.
- Ashori, A. and Nourbaksh, A. Reinforced wood/polypropylene composites: effects of chemical composition and particle size. *Biores. Technol.*, 2010, 101, 2515–2519.
- Renner, K., Kenyo, C., and Moczo, J. Micromechanical deformation processes in PP/wood composites: particle characteristics, adhesion mechanisms. *Compos. Part A- Appl. S.*, 2010, 41, 1653–1661.
- Tabari, H., Nourbakhsh, A., and Ashori, A. Effects on nanoclay and coupling agents on physical-mechanical and water absorption properties of sawdust/polypropylene composites. *Polym. Eng. Sci.*, 2012, 14(1), 123–128.
- Thumm, A. and Dickson, A. The influence of fiber length and damage on mechanical performance of polypropylene/wood pulp composites. *Compos. Part A- Appl. S.*, 2013, 46, 45–62.
- Dos Santos, L. P., Trombetta, E., Flores-Sahagun, T. S., and Satanarayana, K. G. Effect of domestic compatibilizer on performance of polypropylene-sawdust composites. *J. Compos. Mater.*, 2016, 50, 1353–1365.
- Homkiew, C., Ratanawilai, T., and Thongruang, W. Composites from recycled polypropylene and rubber-wood flour: effects of composition on mechanical properties. *J. Thermoplas. Compos. Mater.*, 2015, 28(2), 179–194.
- Ahn, S. H. and Kim, D. S. Effects of recycled PP content on the physical properties of wood/PP composites. *Polymer Korea*, 2014, 38(2), 129–137.
- Seo, Y. W. and Kim, D. S. Effects of wood flour size on the physical properties of polypropylene/wood flour composites. *Polymer Korea*, 2014, 38(3), 327–332.
- Soccalingame, L., Bourmand, A., and Rerrin, D. Reprocessing of wood flour reinforced polypropylene composites. Impact of particle size and coupling agent on composite and particle properties. *Polymer Degradation and Stability*, 2015, 113, 72–85.
- Pereira, L., Dos Santos, L. P., and Flores-Sahagun, T. S. Effect of processing parameters on the properties of polyethylene-sawdust composites. *J. Compos. Mater.*, 2015, 49, 3727–3740.
- Xie, L., Guvenerberg, T., Stenernagel, L., and Ziegmann, G. Influence of particle concentration and type on flow, thermal and mechanical properties of wood-polypropylene composites. *J. Reinf. Plast. Compos.*, 2010, 29, 1940–1951.
- Cui, Y.-H., Wang, X.-X., Xu, Q., and Xia, Z.-Z. Research of moisture absorption behaviour of recycled polypropylene matrix wood plastic composites. *J. Thermoplast. Compos. Mater.*, 2011, 24, 65–68.

Polüpropüleenist ja kasevineeri lihvimistolmusest valmistatud puitplastkomposiitide kasutusomadused

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Paljud teadlased on looduslike biokomposiitide tootmise ja uuringutega tihedalt seotud. Nendes biokomposiitides on võimalik kasutada mitmeid erinevaid polümeere, maatriksina peamiselt polüolefiine ja sarrusena looduslikke kiude või nende kombinatsioone. Osa teadlasi uurib puidutööstuse jääkide kasutamist puitplastkomposiitides (WPC). Antud töös on keskendunud kasevineeri tootmise jääkproduktist, milleks on lihvimistolm (PSD), ja maatriksina polüpropüleenist (PP) valmistatud puitplastkomposiitide kasutamismaduste (tõmbetugevus, paindetugevus ja elastsusmoodul, löögitugevus, mikrokõvadus ning veeimavus) uurimisele. SEM-analüüsiga on uuritud ka komposiitide voolavust ja purunemist. Puitplastkomposiitide modifitseerimiseks kasutati maleiinhappe anhüdriidiga modifitseeritud polüpropüleenit (MAPP). Tulemused näitasid, et PSD ja MAPP-i kasutamine polüpropüleeniga valmistatud WPC-l andis positiivseid tulemusi.