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Wood-Polypropylene Composites: Influence of Processing on the Particle Shape and Size in Correlation with the Mechanical Properties Using Dynamic Image Analysis

The particle size and shape are of momentous significance for the mechanical properties of plastic composites. However, natural fillers, like wood, are not consistent in these attributes. In order to investigate the shared traits between these characteristics, WPCs were produced using polypropylene, hardwood and softwood fillers with different particle sizes and a coupling agent. Afterwards, specimens were processed using an injection molding machine. The filler sizes and shapes were measured using dynamic image analysis. Furthermore, a shortening of coarser particles was detected. Mechanical tests were carried out to gain information about the tensile, flexural and Charpy impact properties. Neither very coarse nor very fine particles showed the best results. Instead, medium-sized particles proved to be the best option. The evaluation of the particle geometry verified a definite correlation between the shape and the mechanical properties, especially regarding the convexity, which can be a useful indicator of the quality of fiber-matrix interaction. The fiber orientation in the matrix was visualized with images taken by X-ray micro tomography.

1 Introduction

In the last 20 years, wood-plastic composites (WPC) gained significance as a commercial material, especially in building and construction industries. Also, in more technically oriented industries, like the automotive sector, the market share of WPC is increasing (Oksman and Sain, 2008). The used fillers usually consisted of wood fibers or wood flours with different sizes and shapes, and were taken from different plant sources, e.g. hardwood or softwood, which were separated and characterized using a sieving process. These products usually contain very large variations of particle lengths and often the specifica-

tions of the manufacturers strongly deviate from the actual sizes of these particles (Stark and Berger, 1997). While the size and shape of these fillers are important for the mechanical properties of WPC, they are crucial regarding the reliability of such materials (Clemons, 2008). Furthermore, the properties of natural fillers may vary from year to year depending on the supplier, growing area, or type (Bradow and Davidonis, 2000; Oksman et al., 2003; Beta and Corke, 2001).

Previous research focused on various points, including the mechanical properties and the influence of the particle length and shape (Gozdecki et al., 2011: Stark, 1999: Stark and Rowlands, 2007; Fasihi and Garmabi, 2011; Leu et al., 2012; Nourbakhsh et al., 2010; Bouafif et al., 2009; Bledzki and Faruk, 2003). Some of these research studies show different results concerning the effect of the particle size on the mechanical properties. On the one hand, the increasing particle size results in an increase in properties like the tensile strength. On the other hand, a decreasing particle size results in an increase of these properties (Leu et al., 2012; Nourbakhsh et al., 2010; Bouafif et al., 2009; Chen et al., 2006; Zaini et al., 1996; Salemane and Luyt, 2006). Other investigations show a huge influence of the particle shape, like the aspect ratio, on the mechanical properties (Migneault et al., 2008; Stark and Rowlands, 2007; Gozdecki et al., 2015; Bledzki and Faruk, 2003). Commercial WPC are usually manufactured using wood flour, meaning their L/D (length to diameter) ratios are very small compared to those of composites with fiber reinforcements. Some papers have shown that this leads to a decreased strength compared to the neat polymer because of stress concentrations in the composite (Migneault et al., 2009; Wolcott and Englund, 1999). However, it is also stated that the use of a coupling agent improves the properties of WPCs, which was documented by several authors. Usually a small amount of coupling agent is sufficient to provide better fiber matrix interaction (Ganster et al., 2006; Dányádi et al., 2007; Bledzki et al., 1998).

Nevertheless, most of these research studies were focused on the particle size before processing, ignoring the change in particle size caused by compounding and injection molding. This aspect is included by Teuber et al., who observed shortenings

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up to over 90% depending on the filler, wood content and processing conditions (Teuber et al., 2016a; 2016b; 2013). Furthermore, Bouafif et al. (2010) found out that the major part of fiber degradation occurs during the compounding process resulting in a vanishing of very long particles and a significant damaging of particles longer than 1.2 mm. This illustrates the dependency of the results of other research studies on the selected processing conditions. They do not provide any information about the degree of particle shortening.

Many methods are worth considering for analyzing the particle size or shape. An important method is sieve fraction analysis, because this method is used in industrial wood flour production (Specht, 2007). Particle samples pass through sieves with different mesh sizes to be distributed into classes. In doing so, this system provides information about the diameter but not the shape of the particles. This is very crucial in the case of samples that include longer fiber-shaped particles that can pass through mesh sizes smaller than their real length, which, in turn, can lead to incorrect results (Bledzki et al., 2005). An evaluation of sieve fraction analysis always provides massweighted (Q3) size distributions, because all fractions are weighted and calculated by mass. Furthermore, particle size distributions can be number-weighted (O0), length-weighted (Q1) or area-weighted (Q2), whereby median values differ very strongly due to an overestimation of very fine or very long particles.

Another possibility that can be used to determine particle size and also shape is dynamic image analysis (ISO 13322-2 2006). In this method, particles and fibers are measured in motion (moving in a liquid or an air stream), making it possible to measure samples very quickly and nearly automatically (Witt et al., 2004; Yu and Hancock, 2008; Rabinski and Thomas, 2004; Feldmann, 2016). The various diameter, length and shape parameters can be calculated to represent the size distribution of a sample. Califice et al. (2013) showed that the results strongly depend upon the method used and the selected evaluation parameter. When analyzing wood flours or other similar natural products, it was determined that a lengthweighted distribution is the most suitable for this application (Teuber et al., 2016a; Bouafif et al., 2010).

The characterization in this publication focuses on two main points. On the one hand, the mechanical properties such as the tensile, flexural and Charpy impact properties of produced specimen were examined. On the other hand, dynamic image analysis was carried out to measure the particle sizes and different shape factors before and after processing. The results of both methods were compared with each other to detect correlations between several of these properties.

2 Experimental

2.1 Materials

A commonly used polypropylene, PP 575P, whose data is given in Table 1, was obtained from SABIC, Riad, Saudi-Arabia, and used as the polymer matrix. This polypropylene is well-suited for wood-filled applications owing to its low melting point. It can be processed at relatively low temperatures, thus enabling thermal degradation of the wood fillers to be avoided.

Wood particles of different sizes, shapes, and from different plant sources, which were provided by the company JRS, J. Rettenmaier & Söhne GmbH & Co. KG, Rosenberg, Germany, were used as fillers. Table 2 shows the selected properties of the used wood materials. The eight different types were derived from hardwood (beech) and softwood (a mixture of spruce and others). The particle length varied between 20 μ m and around 1000 μ m for each plant source. For a better readability all materials are grouped into the classifications very coarse, coarse, fine, and very fine according to Table 2. The types Arbocel and Lignocel underwent different production processes.

In order to improve the adhesion between the two components and thus, enhance the mechanical properties of the compounds, a coupling agent was used. Licocene PP MA 6452 (fine grain) is a functionalized metallocene polypropylene wax and was provided by the company Clariant AG, Muttenz, Switzerland.

2.2 Processing

The ZSE18 HPe twin screw extruder (Leistritz AG, Nuremberg, Germany) was used to prepare the wood fiber reinforced granules for the injection molding process. The screw diameter was 18 mm with a length-to-diameter ratio of 40 (screw length 720 mm). The system was fed by three gravimetric feeders

	Value	Unit
Melt flow rate (at 230 °C, 2.16 kg load) Density (at 23 °C) Tensile strength (at yield) Tensile elongation (at yield) Flexural modulus Notched impact strength (at 23 °C)	11 905 35 11 1600 22	g/10 min kg/m ³ MPa % MPa J/m
Rockwell hardness (R-scale) Vicat softening point Heat deflection temperature (at 455 kPa)	104 153 98	°C °C

Table 1. Properties of the matrix polymer PP 575P made by SABIC, Riad, Saudi Arabia

(Brabender Technologie GmbH & Co. KG, Duisburg, Germany) designated for the polymer, filler and coupling agent to achieve exact mixing ratios with a throughput of 5 kg h⁻¹. Polymer and coupling agent were added in the main feeder, fillers in a side feeder at 20 D (which equals 360 mm of the screw length). The temperature during the whole process was $180 \,^{\circ}$ C. Before compounding, all wood fillers were dried for 24 h at 103 $^{\circ}$ C, so that a residual moisture content of 0.3 to 0.5% could be achieved. The modular screw configuration was built equivalent to the configuration C2 used by Feldmann et al. (2016) to avoid substantial shearing after the side feeder, thus reducing damage to the fillers during the compounding process. The strand was cooled using compressed air cooling and was pelletized into granules with a length of 3 mm.

The composites were produced with varying wood contents of 30 wt.-% and 40 wt.-% and without and with 5% MAPP, respectively, proportional to the wood content. An overview of all manufactured compounds is given in Table 3.

Type 1a test specimens were made according to DIN EN ISO 527. The employed injection molding machine was an All-rounder 320C Golden Edition, Arburg GmbH + Co. KG, Lossburg, Germany, with a clamping force of 500 kN, a standard

three section screw with a diameter of 25 mm and an open machine nozzle. All compounds were dried at 80 °C for 24 h before processing to reach a remaining moisture content of 0.1 wt.%. A temperature profile ranging from 160 °C to 180 °C was used for all produced compounds to avoid significant thermal degradation of the wood particles. The injection pressure was varied between 480 bar and 650 bar according to the respective filler content and its particle size in order to obtain an internal mold pressure of 400 bar at the end of the flow path.

2.3 Characterization

All processed samples were conditioned at 23 °C and in a relative humidity of 50%. The tensile tests were carried out according to EN ISO 527 using a Z010 universal test machine (Zwick Roell AG, Ulm, Germany) with five samples of type 1A. The evaluated parameters were the tensile strength, tensile modulus, and elongation. In the case of the flexural and Charpy impact tests, the samples were cut to remove the shoulders to receive a bar geometry of $80 \times 10 \times 4$ mm³.

	Classification of particle size	Average particle size µm	Bulk weight μm
Hardwoods			
Lignocel HB 500-1000	very coarse	500-1000	205-305
Lignocel HBS 150-500	coarse	200-400	190-270
Lignocel HB 120	fine	40-120	170-230
Arbocel HW 630 PU	very fine	20-40	200-300
Softwoods			
Lignocel 9	very coarse	800-1100	110-190
Arbocel C 320	coarse	200-500	160-240
Arbocel C 750 FP	fine	40-70	120-170
Arbocel CW 630 PU	very fine	20-40	180-210

Table 2. Different wood fillers with their related sizes and weights (according to the data sheet)

Filler type and size	Filler content wt%	MAPP content (relative to filler content) wt%	Formulation (matrix/filler/MAPP) wt%
Hardwood (4 sizes from coarse to fine*)	30 40	0 5 0 5	70/30/0 68.5/30/1.5 60/40/0 58/40/2
Softwood (4 sizes from coarse to fine*)	30 40	0 5 0 5	70/30/0 68.5/30/1.5 60/40/0 58/40/2

* compare with Table 2 for detailed information on the materials

Table 3. Material compositions of tested wood-plastic composites, MAPP content in relation to the regarding filler content

Three-point bending tests were carried out according to EN ISO 178 to determine the flexural properties. Again, five samples were tested to obtain characteristic values for the flexural strength, flexural modulus, and elongation-at-break. The Charpy impact tests were carried out according to EN ISO 179 using a Zwick Charpy impact machine with a 5 J hammer. Ten unnotched samples were tested to gain information about absorbed energy and, therefore, the Charpy impact strength.

The particle sizes of all composites were measured before processing, after compounding and after injection molding. To separate the wood particles from the polypropylene matrix, a simplified Soxhlet extraction was used. About 10 g of the sample was inserted in boiling xylene for 5 h. So as to rule out any influences of the preparation method, this was carried out on the unprocessed particles as well. The dynamic image analysis system Qicpic (Sympatec GmbH, Clausthal-Zellerfeld, Germany) was used in combination with the liquid dispersion unit Mixcel to measure the particle sizes and shapes. The dispersion unit provides a constant flow of a liquid which contains the sample particles. This flow passes a cuvette with a window, where a high-speed camera captures images of each particle. Afterwards, the software Windox calculates the various parameters of the measured particles. Depending on the objective used, the resolutions of this method were 4.2 µm (objective M7) and 1.1 µm (objective M4). In accordance with ISO norms and in order to increase the accuracy of shape recognition of smaller particles, the provided results only include particles bigger than 100 µm for coarse materials (M7) and 10 µm for fine materials (M4).

Different parameters can be used to describe the size and shape of particles when evaluating them. Using Feret's diameter is the easiest and most accurate way to describe wood particle lengths and widths. It refers to distance of two tangents to the contour of the particle in a well-defined orientation (Yu and Hancock, 2008). The length of the particle, the maximum of this diameter after consideration of all possible orientations, is called Feret_{max} and the width of the particle, the minimum of this diameter is called Feret_{min}. The evaluated shape parameters included the aspect ratio, the sphericity and the convexity, which were calculated using values ranging from 0 to 1. A perfect sphere or circle would have the value 1 for all three parameters. The aspect ratio is the ratio of Feret_{min} to Feret_{max}. A fiber of infinite length would provide an aspect ratio of 0, whereas a value of 1 would be calculated for a cubic particle with same length and width. The convexity describes the compactness of a particle and is defined by the ratio of the projection area of the particle and the area of its convex hull. A convexity of 1 would be a particle with no concave regions. Sphericity is the ratio of the perimeter of an equivalent circle to the real perimeter. Again, the value can be a number between 0 and 1, which indicates the irregularity of the shape. To simplify the illustration of shape parameters, only the results for materials after processing are given in this paper.

Another important point regarding particle length distributions is the type of quantity on which the distribution is based. Distributions can be number-based (Q0), length-based (Q1), area-based (Q2) and volume-based (Q3). Teuber et al. (2016a) showed the difference between all four quantities, and determined that the length-based distributions were well-suited for representing broad-sized distributions with fine and coarse amounts. In addition to the mean, the 10^{th} (x_{10}), 50^{th} (x_{50} or median) and 90^{th} (x_{90}) percentiles were analyzed to reflect these broad-sized distributions even better.

An X-ray micro tomography was carried out to characterize the fiber dispersion in the matrix. The scans were made using a ZEISS Xradia 520 Versa X-ray microscope at a voltage of 50 kV and a current of 79 μ A. With an exposure time of two seconds, 1601 radiographs were captured with a resulting voxel size of 9.88 μ m. This voxel size in combination with a detector resolution of 1024 × 1024 pixels allows a field of view of about 10 mm, which is the full cross section of the sample. The images were reconstructed using TXM Reconstructor Software (Carl Zeiss AG, Oberkochen, Germany). Additionally, the interface between fiber and matrix was investigated, using the scanning electron microscope (SEM) CamScan MV2300 (formerly Electron Optics, now Applied Beams, Beaverton, USA).

3 Results and Discussion

3.1 Particle Characterization

The complete results of all particle length measurements are shown in Table 4. Compared to the estimated mean lengths based on the manufacturer's data sheet (Table 2), all four wood types are significantly larger than expected. This can be attributed to the fact that these materials were produced using a sieving process, which allows long but thin fiber-shaped particles to pass through the sieves that have smaller mesh sizes. Due to the limits of the dynamic image analysis the 10th percentiles are nearly identical for all samples measured with the same objective. Nevertheless, the median or mean and the 90th percentile show the influence of processing very clearly. This data is visualized in Fig. 1. The provided data points are the mean values of the particle size distribution and the bars represent the range between the 10th and 90th percentiles. Especially for the very coarse wood particles, the influence of the compounding process is obvious. The influence of the injection molding process is also present, but rather small compared to the compounding process. The shortening of fiber lengths ranges between 31% and 68% for these compounds. The final average particle sizes of all four materials were within a very small range of 419 µm to 517 µm, which proves that there are not big differences in the resulting average particle sizes. Nevertheless, the 90th percentile indicates that the coarser materials still include particles of 1 000 µm and larger. The fine and very fine particles demonstrate a different result, showing an increase in particle lengths after processing, which is very illogical. It can be attributed to the agglomeration of the very fine particles during the extraction process. Finally, these values do not differ significantly before and after processing. Furthermore, the estimated mean lengths according to the data sheet are very close to the measured values.

The results of geometry analysis are given in Table 5. To simplify the results only the median value of the evaluated shape parameter is given. Contrary to the expectations the difference between materials, especially regarding aspect ratio, is rather small. Nevertheless, sphericity and convexity show

Material	Processing condition	Particle length μm				
		× 10	× 50	× 90	Mean	Appreciated mean length
HB 500-1000	(0)	319	1517	2 589	1 540	750
hardwood	(1)	157	639	1 550	763	
very coarse	(2)	116	311	1 162	493	
Lignocel 9	(0)	120	1041	2 789	1 193	950
softwood	(1)	144	469	1 357	640	
very coarse	(2)	120	280	1 287	517	
HBS 150-500	(0)	141	916	1 460	956	300
hardwood	(1)	197	632	1 278	665	
coarse	(2)	118	294	1 052	433	
C 320	(0)	143	624	1 003	609	350
softwood	(1)	133	418	807	446	
coarse	(2)	131	357	799	419	
HB 120	(0)	12	27	147	64	80
hardwood	(1)	12	23	134	55	
fine	(2)	13	36	125	59	
C 750 FP	(0)	12	27	143	59	55
softwood	(1)	12	29	136	56	
fine	(2)	14	45	141	63	
HW 630 PU	(0)	12	22	63	31	30
hardwood	(1)	12	25	67	33	
very fine	(2)	13	34	78	40	
CW 630 PU	(0)	12	24	73	35	30
softwood	(1)	12	25	67	33	
very fine	(2)	13	32	78	39	

Table 4. Results of the particle size measurements before processing (0), after compounding (1) and after injection molding (2) (appreciated mean length based on the data sheet)



Fig. 1. Measured particle sizes of all wood materials. The numbers below the graph indicate the processing status (unprocessed (0), compounded (1), injection molded (2). The provided values are medians (x50), bars indicate the 10th and 90th percentiles (x10, x90)

significant differences. The lowest values for all shape parameters were achieved by both fine materials HB 120 and C 750 FP. The coarser materials tend to have higher values and therefore can be identified as more spherical.

Another important point is the fiber dispersion which has not yet been considered. All measurements of particles after processing where made using an extraction process, which gives no information about the fiber orientation in the sample. Figure 2 shows images taken by X-ray micro tomography of the softwood materials to point out the major differences regarding fiber orientation between the different particle sizes. The image shows the cross-sections of three tensile specimen in a three-dimensional visualization with their respective topdown views (A) and frontal views (B). Big differences can be seen regarding the included voids which are relatively large in the very coarse material. Additionally, the bigger par-

ticles do not show such a homogenous distribution as the fine particles. A big deviation regarding the average particle size can be seen, too. The coarse material shows the typical fiber orientation of an injection molded sample with a strong orientation in the flow direction of fibers which are close to the cavity wall. The fine material shows a very good dispersion with no agglomerates and some few voids that are rather small. This could be an explanation for the difference in the resulting properties. Additionally, Fig. 3 shows images of fractured surfaces of compounds with different particle sizes taken with scanning electron microscopy with a magnification of 500×. The micrographs depict the interface characteristics between fiber and matrix and illustrate that the efficiency of the coupling agent is even better with smaller particles. The image of the very coarse material shows surfaces of wood particles, which are not completely wetted by polymer. Smaller

	Aspect ratio	Sphericity	Convexity
HB 500-1000	0.38	0.54	0.81
HBS 150-500	0.36	0.53	0.80
HB 120	0.31	0.35	0.62
HW 630 PU	0.42	0.49	0.67
Lignocel 9	0.36	0.50	0.81
C 320	0.34	0.49	0.73
C 750 FP	0.29	0.33	0.56
CW 630 PU	0.39	0.40	0.59

Table 5. Median values of the evaluated shape parameters (dimensionless)



Fig. 2. X-ray micro tomography tensile samples with very coarse (1), coarse (2) and fine (3) particles. The additional images show the respective top-down view (A) and frontal view (B)



Fig. 3. SEM micrographs of fractured surfaces with very coarse (1), coarse (2) and fine (3) particles showing the fiber-matrix interface

particles tend to disperse very well and are completely covered by the matrix material.

3.2 Tensile Properties

All tested samples displayed a similar stress-strain behavior with their elongations ranging from 1.2% to 5.1% and their tensile strengths ranging from 24.2 MPa to 44.6 MPa. The tensile modulus ranges from 2800 MPa to 4800 MPa. Figure 4 shows the stress-strain curves of selected composites that represent the lower and upper range of the evaluated properties.

To summarize the influences of the different factors, the main effects on the tensile strength are shown in Fig. 5. As expected, the coupling agent significantly influences the tensile strength due to the improved matrix-filler interaction. The influence of the particle size was very noticeable too, with a local maximum having been achieved by the fine particles and the smallest values having been obtained for very coarse particles. Furthermore, the wood type has a minor effect, and the softwood materials tend to achieve slightly better results. The filler content shows no significant effect in this chart due to an interaction with the coupling agent. This means that higher filler content reduces mechanical properties in the absence of coupling agent, whereas the coupling agent helps produce improved properties as the filler content increases up to a certain point. This indicates the suitability of the coupling agent, which enables even better filler-matrix interaction with a higher filler content. Higher strengths were obtained while using a coupling agent than in neat polypropylene. This contradicts some of the statements made by other researchers (Migneault et al., 2009; Wolcott and Englund, 1999).



Fig. 4. Stress-strain curves of selected composites (HB 500-1000 is a very coarse hardwood; C 750 FP is a fine softwood)



Fig. 5. Main effects of tensile strength. The dotted lines depict the 95% confidence intervals of the given data

The detailed results for the tensile strength and modulus can be seen in Fig. 6. The chart clarifies the negative influence of very coarse materials on the mechanical properties. Additionally, very fine materials seem to reduce the tensile strength as well. The best results were obtained using the medium-sized particles. Furthermore, the composites made of softwoods achieved slightly better results than those made of hardwoods. The tensile modulus did not display such a clear trend. It is evident that the very coarse and very fine particles reached lower tensile modulus values than the medium sized particles. Furthermore, the influence of the coupling agent was very small. Increasing the filler content raises the tensile modulus significantly, as correctly described by Gozdecki et al. (2015).

3.3 Flexural Properties

The results of the bending tests are comparable to the results of the tensile tests. The effects of the different factors are very similar, and show the same tendencies. The elongation ranges from 2.25% to 4.96% and the flexural strength from 50.74 MPa to 81.56 MPa. The flexural modulus was between 2900 MPa and 5100 MPa. The corresponding graphs are depicted in Fig. 7.



Wood type and particle size

Fig. 6. Tensile strength and modulus of all composites with 30% filler content



Fig. 7. Flexural strength and modulus of all composites with 30% filler content

3.3.1 Charpy Impact Properties

Again, a chart depicting the main effects has been provided in Fig. 8 to summarize the influencing factors on the Charpy impact strength. In contrast to the tensile strength, the most significant effect here is the particle size, which displayed enhanced properties for finer particles. This results from an increased notch effect caused by coarser particles, which can act as notches by themselves, weakening the strength of the compound. Additionally, the presence of coarser particles results in a more inhomogeneous distribution and the emergence of larger voids during the injection molding process, which has been shown by means of x-ray images (Fig. 2). Also, as predicted, the addition of coupling agent improved the Charpy properties, while raising the filler content reduced them. This is underlined by the detailed results provided for hardwood materials in Fig. 9.

3.3.2 Correlation of Particle Geometry and Mechanical Properties

Hardwood

When comparing the results for mechanical properties, e.g. the tensile strength, with the obtained particle sizes after processing, it becomes clear that especially large particles, which



Wood type and particle size



Fig. 8. Main effects of Charpy impact strength. The dotted lines depict the 95% confidence intervals of the given data



Fig. 9. Charpy impact strength of all composites with hardwood fillers

are larger than approximately $1\,000\,\mu$ m, affect these properties strongly. In contrast, fillers without particle sizes larger than 80 µm display decreasing strengths as well. To correlate all mechanical properties with particle shape a linear regression fit was carried out. Table 6 shows the correlation coefficients of the tested mechanical properties and geometry parameters. It displays an influence of particle size on all parameters. With declining particle size the moduli decrease while the strengths increase. The correlations of tensile and flexural strength are nearly equivalent which applies for tensile and flexural modulus, too. The moduli correlate only significantly with aspect ratio, but not with sphericity and convexity, which means longer particles result in a higher stiffness. The tensile and flexural strength is highly dependent on sphericity and convexity.

To take also non-linear material behavior and interaction effects between the shape parameters into consideration, a regression analysis was carried out, the results being shown with contour plots in Fig. 10. The calculated tensile strength is displayed with a color gradient. The measured values by tensile testing are marked with squares. Especially the convexity displays significant interactions with the other shape parameters, e.g. with low aspect ratios a change of convexity does not seem to have any effect on the tensile strength whereas with high aspect ratios this effect exists.

4 Conclusion

This study shows the properties of WPC with PP and different wood fillers. The aim was to evaluate the influence of the particle size and shape on the mechanical properties using dynamic image analysis.

Compounding polypropylene and wood flour was carried out with a twin-screw extruder using different wood flours derived from softwoods and hardwoods. The fiber contents 30 wt.-% and 40 wt.-% were selected and coupling agent amounts of 0% and 5% proportional to wood content were employed.

The shortening of fibers mainly took place during the compounding process, but also during the injection molding. It amounted to only 31.1% for the wood flour C320, and ranged between 54.7% and 68% for the other three coarse materials. The mean of the resulting particle length ranges between 419 μ m and 517 μ m. The shortening of the fine particles was not significant, and could not be proved.

As already confirmed by many authors, the use of coupling agent significantly improves the mechanical properties. An increase in tensile strength by 37.4% and an improvement of the Charpy impact strength by 43.2% were verified when using a coupling agent.

The assumption that the particle length is crucial for the mechanical properties was confirmed. Especially, composites with fractions longer than 1 mm showed a drop in their tensile strengths, whereas the finer particles displayed improved mechanical properties that were induced by more homogeneous particle size distributions without the presence of long particles which was shown with an X-ray micro tomography. The difference between coarse and fine particles amounted up to 33.8 % for the tensile strength and 160.1 % for the Charpy impact strength.

The calculated shape parameters provide good correlations with the mechanical properties, e.g. the aspect ratio with the

	Particle size	Aspect ratio	Sphericity	Convexity
Charpy impact strength	-0,834	0,324	-0,361	-0,672
Tensile modulus	0,520	-0,644	0,002	0,276
Tensile strength	-0,666	-0,482	-0,748	-0,802
Flexural modulus	0,548	-0,689	-0,022	0,302
Flexural strength	-0,623	-0,465	-0,711	-0,753

Table 6. Pearson correlation coefficient of mechanical properties and size and shape parameters of batches with 5% coupling agent and 40% filler content



Fig. 10. Contour plots show the correlation between shape parameters and tensile strength. The colors display the calculated results by regression analysis. The squares represent the real measured data points of tensile testing

moduli and sphericity and convexity with the strengths. Especially low values of convexity indicate irregular shaped particles with a higher specific surface area, which results in a higher strength. Contrary to the expectations, the fine particles showed a slightly better aspect ratio than the coarse particles, which can be attributed to the shortening of larger fractions during processing. This corresponds to other publications that showed a positive influence of the aspect ratio on the mechanical properties.

The presented X-ray images substantiate the results and, additionally, show an influence of the dispersion of particles in the matrix, which is closely linked to the particle size.

References

- Beta, T., Corke, H., "Genetic and Environmental Variation in Sorghum Starch Properties", J. Cereal Sci., **34**, 261–268 (2001), DOI:10.1006/jcrs.2000.0379
- Bledzki, A. K., Faruk, O., "Wood Fibre Reinforced Polypropylene Composites: Effect of Fibre Geometry and Coupling Agent on Physico-Mechanical Properties", Appl. Compos. Mater., **10**, 365–379 (2003), DOI:10.1023/A:1025741100628
- Bledzki, A. K., Letman, M., Viksne, A. and Rence, L., "A Comparison of Compounding Processes and Wood Type for Wood Fibre–PP Composites", Composites Part A, 36, 789–797 (2005), DOI:10.1016/j.compositesa.2004.10.029
- Bledzki, A. K., Reihmane, S. and Gassan, J., "Thermoplastics Reinforced with Wood Fillers: A Literature Review", Polym. Plast. Technol. Eng., 37, 451–468 (1998), DOI:10.1080/03602559808001373

- Bouafif, H., Koubaa, A., Perre, P. and Cloutier, A., "Effects of Composite Processing Methods on Wood Particle Development and Length Distribution: Consequences on Mechanical Properties of Wood-Thermoplastic Composites", Wood Fiber Sci., 42, 62–70 (2010), DOI:10.1016/j.compositesa.2009.06.003
- Bouafif, H., Koubaa, A., Perré, P. and Cloutier, A., "Effects of Fiber Characteristics on the Physical and Mechanical Properties of Wood Plastic Composites", Composites Part A, 40, 1975–1981 (2009), DOI:10.1016/j.compositesa.2009.06.003
- Bradow, J. M., Davidonis, G. H., "Quantitation of Fiber Quality and the Cotton Production-Processing Interface: A Physiologist's Perspective", The Journal of Cotton Science, 4, 34–64 (2000)
- Califice, A., Michel, F., Dislaire, G. and Pirard, E., "Influence of Particle Shape on Size Distribution Measurements by 3D and 2D Image Analyses and Laser Diffraction", Powder Technol., **237**, 67–75 (2013), DOI:10.1016/j.powtec.2013.01.003
- Chen, H. C., Chen, T. Y. and Hsu, C. H., "Effects of Wood Particle Size and Mixing Ratios of HDPE on the Properties of the Composites", Holz als Roh- und Werkstoff, **64**, 172–177 (2006), DOI:10.1007/s00107-005-0072-x
- Clemons, C., "Chapter 1 Raw Materials for Wood-Polymer Composites", in Wood-Polymer Composites, Oksman, K., Sain, M. (Eds.), CRC Press, Woodhead Publishing, Boca Raton, Cambridge, UK, p. 1–22 (2008), PMid:21197289; DOI:10.1201/9781439832639.ch1
- Dányádi, L., Janecska, T., Szabó, Z., Nagy, G., Móczó, J. and Pukánszky, B., "Wood Flour Filled PP Composites: Compatibilization and Adhesion", Compos. Sci. Technol., 67, 2838–2846 (2007), DOI:10.1016/j.compscitech.2007.01.024
- Fasihi, M., Garmabi, H., "Evaluation and Optimization of the Mechanical Properties of Highly Filled PVC/(Wood Flour) Composites by Using Experimental Design", J. Vinyl Add. Tech., 17, 112–119 (2011), DOI:10.1002/vnl.20264

- Feldmann, M., "The Effects of the Injection Moulding Temperature on the Mechanical Properties and Morphology of Polypropylene Man-Made Cellulose Fibre Composites", Composites Part A, 87, 146– 152 (2016), DOI:10.1016/j.compositesa.2016.04.022
- Feldmann, M., Heim, H.-P. and Zarges, J.-C., "Influence of the Process Parameters on the Mechanical Properties of Engineering Biocomposites Using a Twin-Screw Extruder", Composites Part A, 83, 113–119 (2016),

DOI:10.1016/j.compositesa.2015.03.028

Ganster, J., Fink, H.-P. and Pinnow, M., "High-Tenacity Man-Made Cellulose Fibre Reinforced Thermoplastics–Injection Moulding Compounds with Polypropylene and Alternative Matrices", Composites Part A, 37, 1796–1804 (2006), DOI 10 101(2017).

DOI:10.1016/j.compositesa.2005.09.005

- Gozdecki, C., Wilczyński, A., Kociszewski, M. and Zajchowski, S., "Properties of Wood–Plastic Composites Made of Milled Particleboard and Polypropylene", Eur. J. Wood Wood Prod., 73, 87–95 (2015), DOI:10.1007/S00107-014-0852-2
- Gozdecki, C., Zajchowski, S., Kociszewski, M., Wilczyński, A. and Mirowski, J., "Effect of Wood Particle Size on Mechanical Properties of Industrial Wood Particle-Polyethylene Composites", Polimery, 56, 375–380 (2011)
- ISO 13322-2:2006, "Particle Size Analysis Image Analysis Methods – Part 2: Dynamic Image Analysis Methods", International Organization for Standardization (2006)
- Leu, S.-Y., Yang, T.-H., Lo, S.-F. and Yang, T.-H., "Optimized Material Composition to Improve the Physical and Mechanical Properties of Extruded Wood–Plastic Composites (WPCs)", Constr. Build. Mater., 29, 120–127 (2012), DOI:10.1016/j.conbuildmat.2011.09.013
- Migneault, S., Koubaa, A., Erchiqui, F., Chaala, A., Englund, K., Krause, C. and Wolcott, M. P., "Effect of Fiber Length on Processing and Properties of Extruded Wood-Fiber/HDPE Composites", J. Appl. Polym. Sci., **110**, 1085–1092 (2008), DOI:10.1002/app.28720
- Migneault, S., Koubaa, A., Erchiqui, F., Chaala, A., Englund, K. and Wolcott, M. P., "Effects of Processing Method and Fiber Size on the Structure and Properties of Wood–Plastic Composites", Composites Part A, 40, 80–85 (2009), DOI:10.1016/j.compositesa.2008.10.004
- Nourbakhsh, A., Karegarfard, A. and Ashori, A., "Effects of Particle Size and Coupling Agent Concentration on Mechanical Properties of Particulate-filled Polymer Composites", J. Thermoplast. Compos. Mater., 23, 169–174 (2010), DOI:10.1177/0892705709340962
- Oksman, K., Sain, M., "Introduction", in Wood-Polymer Composites, Oksman, K., Sain, M. (Eds.), CRC Press, Woodhead Publishing, Boca Raton, Cambridge, UK (2008), DOI:10.1201/9781439832639
- Oksman, K., Skrifvars, M. and Selin, J.-F., "Natural Fibres as Reinforcement in Polylactic Acid (PLA) Composites", Composites Science and Technology, 63, 1317–1324 (2003), DOI:10.1016/S0266-3538(03)00103-9
- Rabinski, G., Thomas, D., "Dynamic Digital Image Analysis: Emerging Technology for Particle Characterization", Nano and Micro Particles in Water and Wastewater Treatment, **50**, 19–26 (2004), DOI:10.2166/wst.2004.0691
- Salemane, M. G., Luyt, A. S., "Thermal and Mechanical Properties Of Polypropylene–Wood Powder Composites", J. Appl. Polym. Sci., 100, 4173–4180 (2006), DOI:10.1002/app.23521
- Specht, K.: Holz- und hanffaserverstärktes Polypropylen in der Spritzgie
 ßverarbeitung, Faseraufschluss- und Verbundaufbereitungsverfahren, Haftvermittler, Alterungsverhalten, Lehrstuhl Kunststoffund Recyclingtechnik, Kassel (2007)
- Stark, N. M., "Wood Fiber Derived from Scrap Pallets Used in Polypropylene Composites", Forest Products Journal, 49, 39–46 (1999)

- Stark, N. M., Berger, M. J., "Effect of Particle Size on Properties of Wood-Flour Reinforced Polypropylene Composites", The Fourth International Conference on Woodfiber-Plastic Composites, Madison,WI, USA, p. 134–143 (1997)
- Stark, N. M., Rowlands, R. E., "Effects of Wood Fiber Characteristics on Mechanical Properties of Wood/Polypropylene Composites", Wood Fiber Sci., 35, 167–174 (2007)
- Teuber, L., Militz, H. and Krause, A., "Characterisation of the Wood Component of WPC via Dynamic Image Analysis", in "Track A: Materials and Technologies", in "Characterization of Fibres and Particles", Geldermann, J., Schumann, M. (Eds.), First International Conference on Resource Efficiency in Interorganizational Networks, Göttingen, p. 46–54 (2013)
- Teuber, L., Militz, H. and Krause, A., "Dynamic Particle Analysis for the Evaluation of Particle Degradation during Compounding of Wood Plastic Composites", Composites Part A, 84, 464–471 (2016a), DOI:10.1016/j.compositesa.2016.02.028
- Teuber, L., Militz, H. and Krause, A., "Processing of Wood Plastic Composites: The Influence of Feeding Method and Polymer Melt Flow Rate on Particle Degradation", J. Appl. Polym. Sci., 133, 43231 (2016b), DOI:10.1002/app.43231
- Witt, W., Köhler, U. and List, J., "Direct Imaging of Very Fast Particles Opens the Application of Powerful (Dry) Dispersion for Size and Shape Characterisation", PARTEC, Nuremberg (2004)
- Wolcott, M. P., Englund, K., "A Technology Review of Wood-Plastic Composites", 33rd International Particleboard/Composite Materials Symposium, Washington State University, Pullman, p. 103–111 (1999)
- Yu, W., Hancock, B. C., "Evaluation of Dynamic Image Analysis for Characterizing Pharmaceutical Excipient Particles", Int. J. Pharm., 361, 150–157 (2008), PMid:18573322; DOI:10.1016/j.ijpharm.2008.05.025
- Zaini, M. J., Fuad, M. A., Ismail, Z., Mansor, M. S. and Mustafah, J., "The Effect of Filler Content and Size on the Mechanical Properties of Polypropylene/Oil Palm Wood Flour Composites", Polym. Int., 40, 51–55 (1996), DOI:10.1002/(SICI)1097-0126(199605)40:1<51::AID-PI514>3.0.CO;2-I

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