Processing of wood plastic composites: The influence of feeding method and polymer melt flow rate on particle degradation

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ABSTRACT: Spruce wood particle (WP)/polypropylene (PP) compounds were prepared in an internal mixer using different rotor speeds. To analyze the effect of feeding method on particle degradation, WP and PP were either fed as dry-blend or WP was fed into the PP melt. To prevent melt freezing, pre-heated WP were used as comparison to cold WP. In addition, WPs were compounded with different grades of PP or high-density polyethylene (HDPE) to analyze the effect of polymer matrix melt flow rate (MFR) on particle degradation. Mixing behavior of compounds containing 30% and 70% (w/w) WP depended on feeding method, represented by a changing relation of final torque values. Feeding as dry-blend and using pre-heated particles led to stronger WP degradation. Degradation decreased with increasing polymer MFR. For PP compounds, particle degradation was stronger when containing 70% WP, for HDPE the difference due to WP content was only marginal. © 2015 Wiley Periodicals, Inc. J. Appl. Polym. Sci. **2016**, *133*, 43231.

INTRODUCTION

The interest in wood plastic composites (WPC) as partly bio-based materials for automotive, construction, and consumer applications has experienced an increase represented by a recent growth in market share in the European Union.[1] WPC consists of a polymer matrix—commonly polypropylene (PP), polyethylene (PE) or polyvinyl chloride (PVC)—a wood filler in the form of fibers, particles or flour, and additives (e.g., coupling agents, pigments, and UV stabilizers).[2] Due to their thermoplastic nature, it is possible to process WPCs on the same equipment as neat thermoplastics.[3] The material composition and production process have to be chosen and optimized in such a way that the resulting composite properties match the requirements specific to the field of application. One factor that has been identified to influence the composite properties is the size and shape of the filler.[4] For example, particles with their low length to diameter ratio lead to more homogenous materials as they are easier to disperse in the polymeric matrix.[5,6] But compared to particles, fibers have a high length to diameter ratio and therefore better enhance the tensile and flexural strength.[7] Also the rheological properties of a polymer are dependent on filler morphology, making amendments of the compounding process necessary. Polymer melts possess non-Newtonian flow behavior. Their viscosity is not constant but underlies shear-thinning behavior, i.e., it decreases when the shear rate increases.[8] Several studies demonstrated that the polymer viscosity is generally higher when filler is added and it increases with increasing filler content.[3,9–13] Also the degree of shear-thinning becomes stronger with increasing filler content.[11,14] Zhang *et al.*[15] and Yuan *et al.*[16] found out that viscosity is not dependent only on filler content but also on filler size. In their studies, long wood fibers in the composite resulted in higher viscosities whereas a higher amount of short fibers lead to lower composite viscosities. Huber *et al.*[17] made similar observations when wood particles (WPs) were used as filler. Stark and Berger[18] stated an opposite trend for wood fiber filled composites.

However, it has been shown that initial filler morphology is not constant throughout processing, but it is prone to changes due to shear forces and high temperature.[19] The extent of filler degradation in extrusion compounding, for example, was found to depend on process settings like degree of filling, screw design, screw rotation speed, and feed rate.[20–25]

While the influence of filler morphology on polymer properties, e.g., polymer rheology, was studied to a vast extent, there are

Table I. Process Variants

Wood content (wt %)	Feeding method	Rotor speed (rpm)	Preheating
30	s	20	c
		60	c
		120	c, h
	t	20	c
		60	c
		120	c, h
70	s	20	c
		60	c
		120	c, h
	t	20	c
		60	c
		120	c, h

Feeding: s = separately, t = together; pre-heating: c = "cold," h = "hot."

Figure 1. Size distributions of initial particle length (A) and elongation (B) as box-whisker plots. The bottom and top of the box represent the 25% and 75% quartile. The ends of the whiskers represent the 10% and 90% quantile.

only few studies that directly analyze the effect of polymer rheological properties, e.g., polymer melt flow rate (MFR), on wood filler size. Balasuriya et al.[3] compounded wood flakes and HDPE with different MFR to study the effect on flake distribution and wetting by the matrix and to analyze the mechanical properties resulting thereof. Peltola et al.[26] compounded wood fibers with polylactic acid (PLA) and PP and concluded that differences in fiber degradation result from the higher viscosity of the PLA matrix. Ren and Dai[27] found for glass fiber/PP composites that fiber length degradation decreases with increasing MFR of the matrix polymer.

Also the way of feeding the wood filler to the compounding process can play a role in filler degradation. The wood filler is either fed to the process at the same time as the polymer granules, forming a dry-blend before the polymer starts to melt, or the wood is introduced into the already molten polymer.[28] Feeding as a dry-blend simplifies the machine setup as only one infeed is needed. But it can also lead to severe filler size reduction due to increased particle–particle interaction with the unmolten polymer granules. This effect can be overcome by introducing the wood into the molten polymer. In twin-screw

Table II. Polymers Used in the Study

Brand name	MFR (g/10 min)	Sample name
PP 505P	2	PP2.0
PP 575P	11	PP11
PP 579S	47	PP47
HDPE CC252	2.2	PE2.2
HDPE 0863F	8	PE8
HDPE CC3054	30	PE30

All polymers obtained from SABIC. MFR as specified in the product data sheets and determined at 230°C and 2.16 kg for PP and at 190°C and 2.16 kg for HDPE.
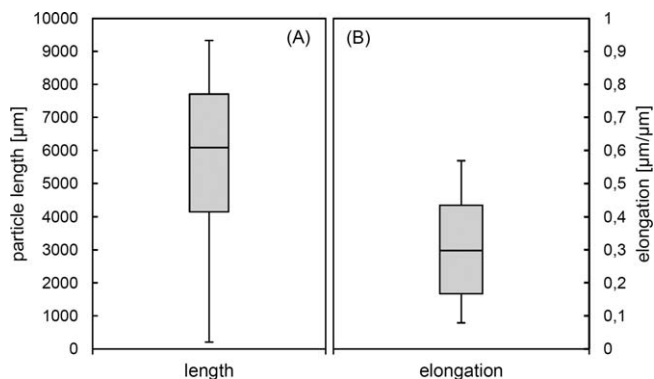
compounding, a side-feeder can be used to introduce the filler further downstream of the extruder. Though, side-feeding can lead to poorer filler dispersion due to a shorter mixing time.[29] When the filler is introduced into the polymer melt, there is a huge temperature difference between the two materials. When the filler comes into contact with the polymer melt, it will primarily be heated by the latter. This leads to a loss in temperature of the melt which will be balanced by the heating of the processing equipment further on. Due to the loss in temperature, the viscosity of the melt will increase.[8] This will increase the shear forces acting on the filler material.[26] When considering a specific heat capacity of 1.35 kJ/(kgK) for spruce wood (at a moisture content of 0%)[30] and 1.7 kJ/(kgK) for PP,[31] and a target filler content of at least 50%, the temperature of the polymer might drop below the melting point causing a local freezing of the melt. The presence of solid polymer can again lead to an increase in filler degradation.[32]

The present study analyzes the dependency of wood filler degradation on feeding method and polymer MFR using an internal mixer, which allows to record torque and temperature development during processing. In the first part of the study, WPs were compounded with PP with varying wood content and rotor speed and by (i) feeding wood and polymer as dry-blend and (ii) feeding the particles into the polymer melt. Also preheating of the WPs was considered. In the second part of the study, WPs were compounded with different grades of PP and HDPE having low, medium and high MRF to study the influence of polymer type and MFR on WP degradation.

EXPERIMENTAL

Wood Particle Production
Norway spruce (*Picea abies*) wood was ground with a cutting mill with aperture size of 4 mm and subsequently screened via sieve separation to a fraction of 1–2 mm particle size. The resulting WPs were washed in water between two sieves with mesh size 0.71 mm and 1.6 mm to remove fines. Afterward, the WPs were dried in a hot air oven to a moisture content of <2%.

Table III. Summary of Results

Wood content (wt %)	Feeding method	Rotor speed (rpm)	Pre-heating	Particle length (μm)			Particle diameter (μm)			Elongation (μm)			Torque (Nm)	Temperature (°C)
				x_{25}	x_{50}	x_{75}	x_{25}	x_{50}	x_{75}	x_{25}	x_{50}	x_{75}		
30	s	20	c	64	125	233	25	35	51	0.260	0.358	0.475	12.5	184
		60	c	53	98	174	24	34	48	0.293	0.390	0.513	14.1	189
		120	c	49	88	154	24	33	46	0.304	0.402	0.520	10.9	192
		120	h	47	84	144	24	33	45	0.308	0.405	0.524	7.3	190
	t	20	c	65	117	199	25	35	50	0.265	0.360	0.474	8.8	183
		60	c	51	90	148	24	33	45	0.298	0.392	0.510	14.1	189
		120	c	49	86	145	24	33	46	0.309	0.405	0.521	15.0	197
		120	h	46	80	136	24	33	46	0.325	0.422	0.548	13.1	196
70	s	20	c	60	114	207	25	35	49	0.270	0.367	0.482	8.4	183
		60	c	46	80	134	24	33	44	0.321	0.415	0.535	12.5	188
		120	c	43	71	117	24	32	43	0.344	0.438	0.562	14.5	196
		120	h	40	64	104	23	32	40	0.354	0.445	0.570	14.5	194
	t	20	c	61	113	197	25	35	50	0.275	0.369	0.485	10.6	183
		60	c	45	76	125	24	33	44	0.328	0.422	0.544	15.6	189
		120	c	40	66	108	23	32	41	0.349	0.442	0.567	15.5	198
		120	h	37	59	96	22	31	39	0.363	0.454	0.579	16.6	198

Feeding: s = separately, t = together; pre-heating: c = "cold," h = "hot."

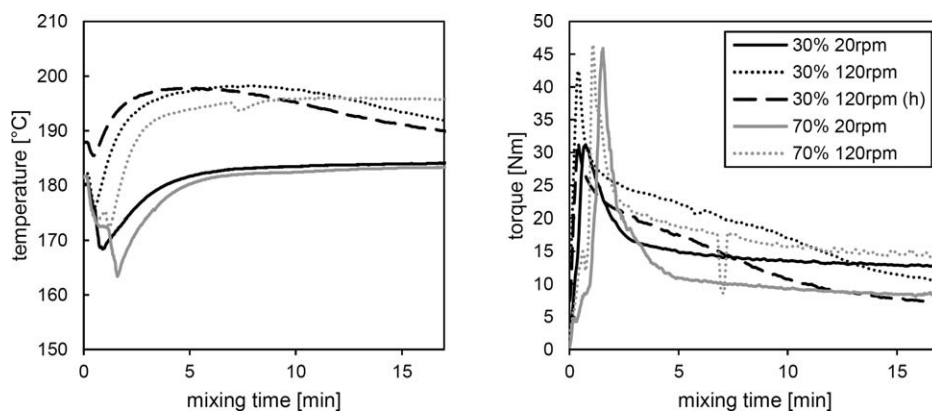**Figure 2.** Progression of temperature and torque for selected process variants after adding WP into the polymer melt (h: preheated particles).

Composite Preparation

Mixing of WP and polymer was done with HAAKE Polylab OS Rheodrive 7 combined with the HAAKE Rheomix OS 3000 (Thermofischer Scientific) laboratory mixer with Banbury rotors. The degree of filling of the free volume was set to 60% and processing temperature was 180°C. Mixer torque and mass temperature were recorded with HAAKE Polysoft OS software version 2.4.0.28.

For the first part of the study, WPs were mixed with PP SABIC PP 575P (Saudi Basic Industries Corporation, Saudi Arabia) at varying wood content (30% and 70% by weight) and rotor speed (20, 60, and 120 rpm). Each run was carried out with two different feeding methods. On the one hand, polymer and WPs were dry-blended prior to mixing and fed to the mixer together. On the other hand, the polymer was mixed until steady torque was reached and the WPs were added to the polymer melt. At a rotor speed of 120 rpm additional runs with WP pre-heated to 140°C were carried out. Details concerning the different process variants are listed in Table I. When the temperature reached 195°C for the first time, the mixer was opened for a few seconds to lower the temperature again to prevent
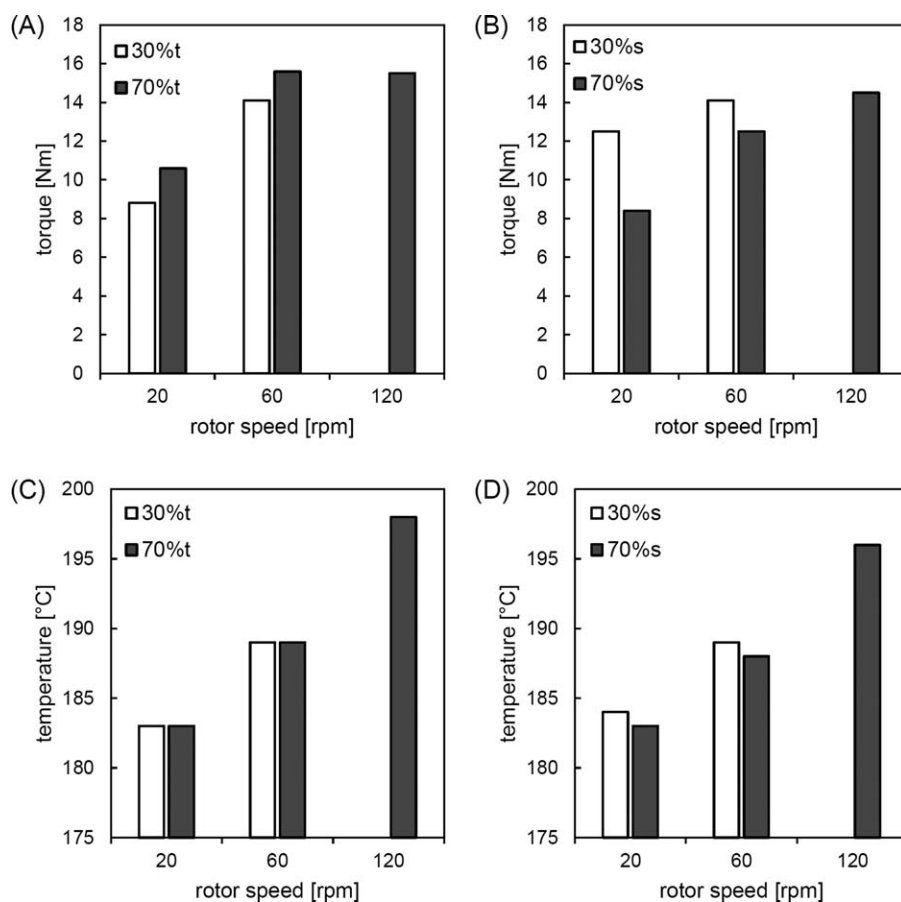

Figure 3. Final torque (A,B) and temperature (C,D) after 16.5 min of mixing when polymer and WP are fed together (A,C) or separately (B,D).
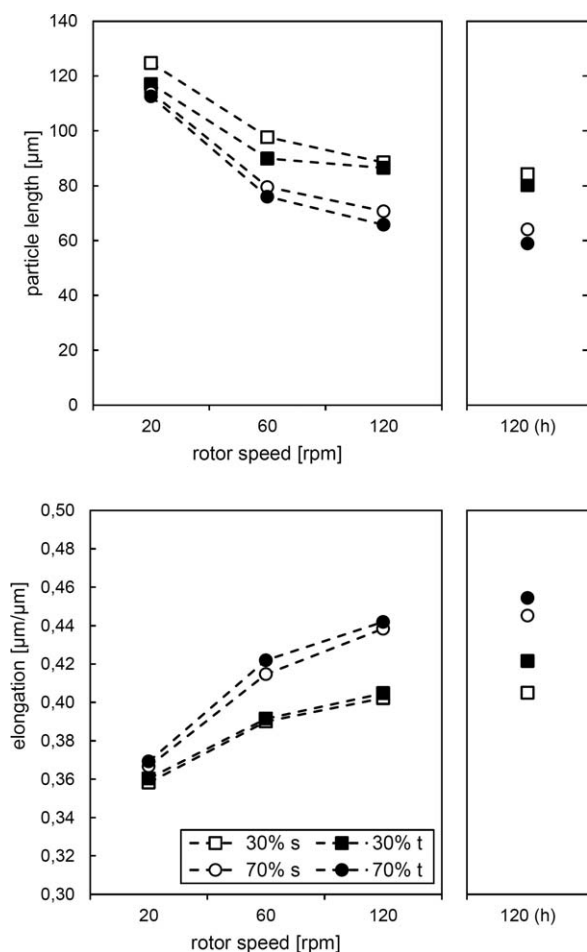
Figure 4. WP length and elongation after mixing (h: preheated particles).

thermal wood degradation. This was necessary only with 70% wood content mixed at 120 rpm. To eliminate the effect of residence time in the mixer on WP degradation, mixing was stopped for both feeding methods 17.2 min after WP introduction and the compound was removed from the mixer to cool down to room temperature.

For the second part of the study, WPs were mixed with different types of PP and HDPE with varying MFR (Table II). All polymers were supplied by SABIC (Saudi Basic Industries Corporation, Saudi Arabia). Rotor speed was 60 rpm. Compounds were produced with a wood content of 30% and 70% by weight. The WPs were fed into the polymer melt without pre-heating. Mixing time was set to 7.5 min after WP introduction to reduce the influence of residence time on particle degradation since the first part of the study showed that mixing conditions for these process settings are already stable after this time.

Particle Extraction and Characterization
To separate the WP from the polymer matrix the compounds were Soxhlet extracted in hot xylene for 8 h. Afterward, the WPs were kept at room temperature for excess solvent to evaporate. To exclude any influence of the extracting method on particle morphology, unprocessed particles underwent the extraction process, too, and their size and shape were compared

to the initial values. WP size and shape were determined via the dynamic image analysis system QICPIC combined with vibrating chute VIBRI and dry dispersion unit RODOS (Sympatec GmbH, Germany). In the QICPIC, the particles are dispersed in an accelerated air jet and a high-speed camera takes pictures of the particle stream. Measuring particle size and shape from the projected particle area of binary pictures and calculating size and shape distributions was done by the software WINDOX (Sympatec GmbH, Germany). Particles before and after processing were measured with a resolution of 20 µm and 10 µm, respectively. Particle length was characterized with respect to the median and quartiles of the length-based particle length distribution (Q1) (DIN ISO 9276-1).[33] Thereby, particle length was defined as the shortest path between the most distant end points of the particle after skeletonizing its projected area.[34] Particle elongation was analyzed with respect to the median and the quartiles of the number-based distribution including only particles with a length of at least 5× resolution (length >100 µm and >50 µm for particles before and after processing, respectively). Elongation was defined as the ratio of particle diameter to particle length with the diameter being the projected area divided by the added length of all skeleton paths. The number of particles analyzed was approximately 6,800 for the particles before processing and varied between 0.5 million and 4 million per composite sample.

RESULTS AND DISCUSSION

Initial Particle Size and Shape
Figure 1 shows the initial particle length and elongation distributions. The particle length distribution was relatively narrow with a median length of 6068 µm and the 25% and 75% quartiles at 4153 µm and 7613 µm. Although the WPs were washed in water to remove dust, they still contained fines represented by a 10% decile at 202 µm. The elongation was ranging from 0.079 to 0.569 with a median of 0.298.

Effects of Process Conditions
A summary of the results representing the effects of process conditions on particle size and shape and final torque and temperature is given in Table III. Figure 2 exemplarily shows the change in temperature and torque during mixing after adding WPs for some process variants, among them the two variants with cold and pre-heated particles containing 30% WP, fed into the polymer melt and processed at 120 rpm. While all processes reached a plateau temperature and torque, the curves for the two variants mentioned above were still sloping at the end of the mixing time, indicating that the mixing process was not completed yet. Due to this deviating mixing behavior, these two variants will be excluded from further analyses with respect to final torque and temperature.

Final Torque and Temperature. As can be seen in Figure 3, torque and temperature increased with increasing rotor speed for both feeding methods. Increasing the rotor speed requires a higher torque from the mixer motor. The increase in torque was not linear with rotor speed but was higher for lower rotor speeds and lower for higher rotor speeds. Due to the shear-thinning behavior of the melt, its resistance to flow decreases when the shear rate, i.e., rotor speed, is increased.[8] In turn, the
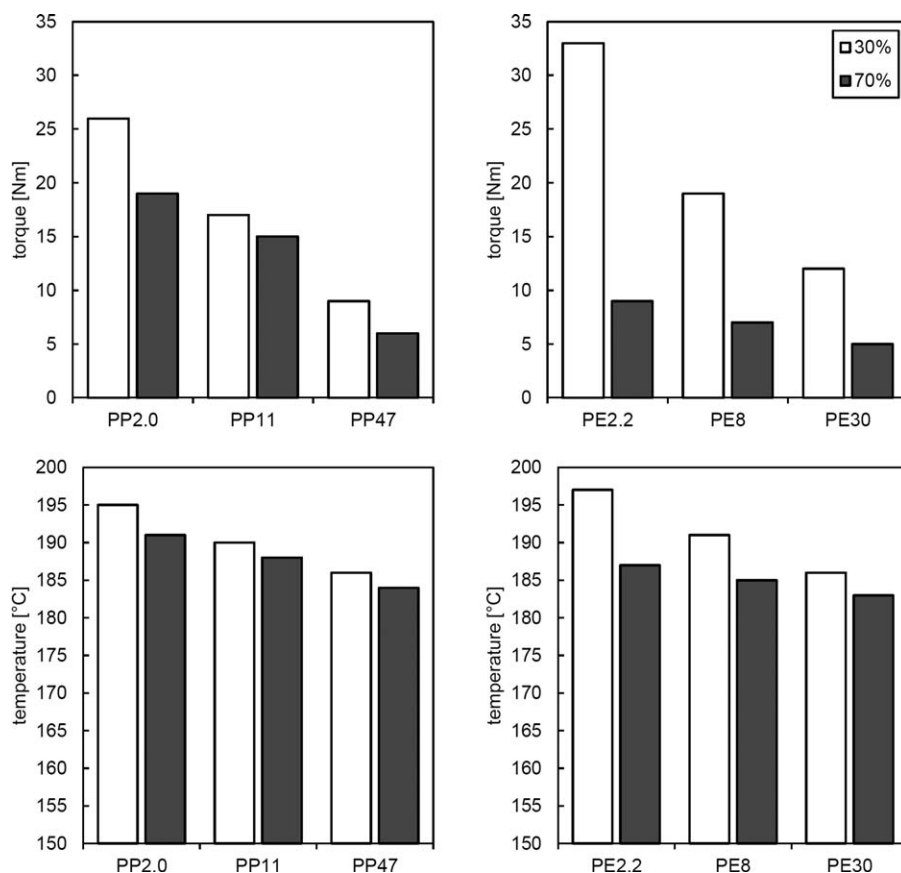
Figure 5. Final torque and temperature after 7.5 min of mixing.

decrease in flow resistance reduces the rate of increase in mixer motor torque to obtain higher rotor speeds. The increase in melt temperature with increasing rotor speed results from the growth of viscous energy dissipation in the polymer.[35]

When WP and PP were fed as dry-blend, the torque was higher for composites containing 70% WP compared to 30% WP (Figure 3A). This can be attributed to the higher viscosity of composites having higher filler contents. At a higher filler concentration, the flow of the polymer is restrained by the particles and particle–particle interaction increases.[10,15]

However, when the WP were fed into the polymer melt, the torque was lower for 70% wood content compared to 30% wood content (Figure 3B). It was even lower than for 70% wood content fed as dry-blend. In the dry-blend, the small fraction of polymer granules was equally distributed between the WPs, providing a uniform wetting of the filler when the polymer started to melt. The rheological behavior of the composite melt and the measurable torque were then related to viscous interactions between the particles.[36] Polymer melts and especially filled polymers are known for their wall-slip behavior, which was also reported for WPC.[37] During mixing they develop a matrix dominated layer close to the processing equipment wall. The viscosity of this layer is lower than the composite melt viscosity and therefore results in slippage of the bulk material.[8,38] In WPC, the thickness and velocity of the slip layer depend on various factors, e.g., wood species, filler size, filler content. But the

mechanisms of slip layer formation are yet not fully understood.[37] In the present study, the wall-slip phenomenon might explain the different mixing behaviors dependent on feeding method. The separate feeding of polymer and WP might have promoted the formation of a slip layer for the compound filled with 70% wood content. When the low fraction of polymer was introduced to the mixer without the filler, the melt was able to spread on the walls of the mixing chamber and the rotors. Adding the high fraction of WP to the low fraction of polymer melt might have led to a nonuniform wetting of the filler thereby maintaining the slip layer at the mixing chamber walls and leading to a lower apparent viscosity. In addition, the nonuniform wetting might have caused the compounds' rheological properties to be more related to solid friction resulting in lower torque values.[36] For the composites containing 30% WP, the amount of polymer was sufficient to ensure uniform filler wetting and similar flow properties independent of feeding method. However, the formation of a slip layer was not verified and a more detailed study would be necessary to analyze the conditions inside the mixer, which is beyond the scope of the present study.

Particle Size and Shape. Figure 4 shows the median particle length and elongation of WP after processing under different process conditions. Median particle length was reduced by more than 98% of initial length regardless of process conditions reaching values between 64 μm and 125 μm. The 25% and 75%
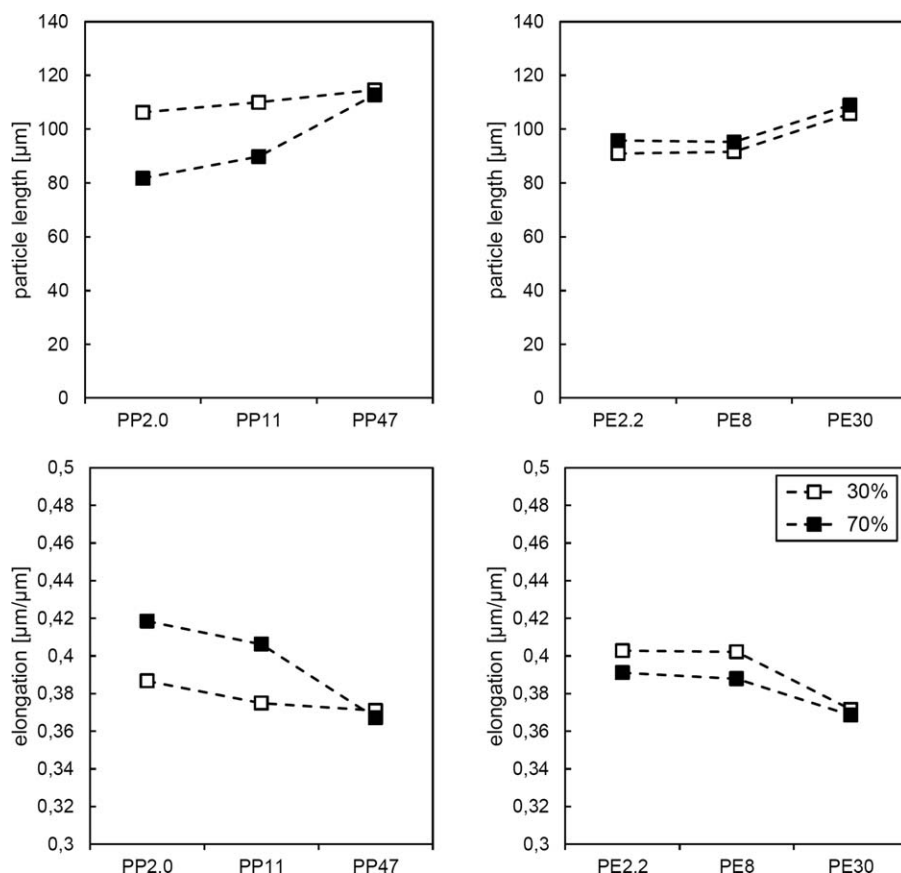
Figure 6. WP length and elongation depending on wood content and polymer matrix MFR.

quartiles of the particle length distribution decreased at a similar rate, i.e., by more than 98% and 96%, respectively. Particle length decreased with increasing rotor speed while elongation increased. Increasing the rotor speed increases the shear forces inside the polymer melt causing the WP to break.[39] In addition, the number of rotations and the distance travelled by the particles inside the mixer increases with increasing rotor speed when mixing time is kept constant. The increase in elongation shows that particle length was more affected by shear than particle diameter.

A strong effect of WP content on particle length and elongation was visible, especially at high rotor speeds. Particles were 1–26% shorter with respect to the median length in composites containing 70% compared to 30% WP due to the stronger particle–particle interaction at high filler contents.[40]

Analyzing the effect of feeding method on particle length revealed that feeding WP and PP as a dry-blend lead to 1–8% shorter particles with respect to the median length compared to feeding WP into the polymer melt. This confirmed the assumption of a milling effect by the solid polymer granules acting like grindstones before they start to soften and melt. This milling effect was overcome when the WPs were fed into the polymer melt and particle length degradation was reduced.

WPs were pre-heated to prevent possible freezing of the polymer melt, thereby attempting to additionally reduce particle degradation. However, the results showed that pre-heating led to stronger particle degradation represented by WP being up to 11% shorter with respect to the median length and having an elongation that was up to 4% higher with respect to the median elongation compared to the cold particles. Pre-heating the WP before processing prolonged their residence time at high temperatures. Wood it is known to experience a significant decrease in mechanical properties when exposed to high temperatures This is caused by the thermal degradation and structural change of cell wall substances, which already starts at temperatures >105°C.[30] In the present study, the loss in WP mechanical strength resulted in a lower resistance of the particles to shear stresses from processing. This effect seemed to have outbalanced the advantage of preventing melt-freezing and led to stronger degradation of the pre-heated WP compared to the cold WP.

Effects of MFI

Final Torque and Temperature. WPs were added into the polymer melt and processed with a rotor speed of 60 rpm to study the influence of polymer type and MRF on final torque and temperature. The overall trends of the results were consistent with the results from the first part of the study for this combination of feeding method and rotor speed. As shown in Figure 5, final torque and temperature were higher for compounds containing 30% WP than for compounds containing 70% WP. For equal filler content, final torque and temperature decreased with increasing MFR for both PP and HDPE compounds. This decrease was caused by a lower composite viscosity when a high

Table IV. Particle Size and Elongation after Process with Different Matrix Polymers

Wood content (wt %)	Polymer	Particle length (µm)			Particle diameter (µm)			Elongation (µm/µm)		
		x_{25}	x_{50}	x_{75}	x_{25}	x_{50}	x_{75}	x_{25}	x_{50}	x_{75}
30	PP2.0	57	106	190	25	35	50	0.291	0.387	0.508
	PP11	58	110	199	25	35	49	0.279	0.375	0.928
	PP47	59	115	213	25	35	49	0.273	0.371	0.491
	PE2.2	51	91	156	24	34	47	0.306	0.403	0.519
	PE8	51	92	159	24	34	47	0.305	0.402	0.523
	PE30	57	106	184	25	34	47	0.277	0.372	0.488
70	PP2.0	48	82	137	24	33	46	0.324	0.419	0.542
	PP11	50	90	155	24	34	47	0.309	0.406	0.524
	PP47	59	113	204	25	34	49	0.271	0.367	0.482
	PE2.2	53	96	163	25	34	47	0.296	0.391	0.511
	PE8	53	95	162	24	34	46	0.292	0.388	0.507
	PE30	58	109	191	25	34	48	0.272	0.369	0.484

MFR matrix polymer is used.[16] An increase in matrix polymer MFR can balance the decrease of composite MFR caused by filler incorporation.[3,16] This effect was also responsible for the torque and temperature values being lower for HDPE compounds compared to PP compounds when both contained 70% WP. The HDPE and PP grades in this study were chosen with respect to their similar range of MFR. But as the MFR of the HDPE grades was determined at a lower temperature than the MFR of the PP grades, their actual MFR during processing was accordingly higher (and their viscosity lower) than that of PP.

However, at a WP content of 30%, torque and temperature were higher for HDPE than for PP compounds. As discussed previously, the low WP fraction was properly wetted by the larger polymer fraction. Filler wetting is even increased with high MFR polymers[3,16,27] and thereby composites resistance to shear is enhanced, leading to a larger relative decrease of composite MFR caused by filler incorporation.[12] Due to the higher actual MFR of HDPE during processing, the effect of increased filler wetting seemed to be dominant for the HDPE compounds with 30% WP content when compared to PP compounds. The resulting higher shear resistance led to higher torque values.

Particle Degradation. The median WP length and elongation after processing and dependent on matrix polymer MFR are represented in Figure 6. The quartiles and the particle diameters are summarized in Table IV. For PP as well as HDPE the particle degradation decreased with increasing MFR. The use of higher MFR matrix polymers lead to lower composite viscosities, which was demonstrated by the lower final torque values. The decrease in viscosity reduced the shear stresses acting on the WP resulting in less severe length reduction.

For the PP compounds, there was an obvious difference in particle length and elongation dependent on wood content. Although the viscosity was apparently higher for 30% filler composites (represented by lower torque values), this was not represented by the particle length results, supporting the assumption of different wall-slip conditions for the present

feeding method. For low and medium MFR, WP were 23% and 18% shorter with respect to the median length when processed at 70% filler content compared to WP processed at 30%. The higher filler content increased the particle–particle interaction and the composite viscosity. When using a high MFR polymer (PP47) resulting particle length was similar for both filler contents. The high MFR matrix polymer must have provided a better wetting of WP due to its enhanced flowability, leading to reduced friction between the WP. This is also represented by the similar elongation for both filler contents with this matrix polymer.

For the HDPE compounds, the difference in resulting particle length dependent on wood content was only marginal with slightly shorter particles when processed at low wood content. Due to its linear structure and high MFR during processing, HDPE can reach high rates of impregnation and penetrate into the lumens and cracks of WP forming a strong physical interlock between filler and matrix.[3,16] This physical interlock between HDPE and WP led to severe particle degradation also at 30% wood content.

CONCLUSIONS

WPC based on WP and either PP or HDPE were compounded in an internal mixer to study the dependency of WP degradation on feeding method and polymer MFR. Although the compounding conditions of an internal mixer are not directly comparable to industrially relevant processes, e.g., twin-screw extrusion, general conclusion can be drawn from the results of the present study that could help to reduce filler degradation during WPC compounding.

The feeding method influenced the final torque when WPs were compounded with PP. The relation of torque values between 30% and 70% filled compounds was reversed when WPs were fed into the polymer melt compared to feeding as dry-blend. This indicated that the mixing behavior of compounds was dependent on feeding method. Regardless of filler content,

feeding as dry-blend led to stronger particle degradation compared to separate feeding, confirming that the polymer granules were acting like grindstones. Pre-heating of WP prior to compounding increased particle degradation. This was explained by the longer residence time at elevated temperatures leading to reduced mechanical strength of the wood. Particle degradation was reduced by using high MFR matrix polymers. While there was a strong dependence of particle length on filler content for PP compounds, the difference was only marginal for HDPE.

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AUTHOR CONTRIBUTIONS

Laura Teuber: conception and design of study, data acquisition, analysis and interpretation of data, drafting manuscript, corresponding author. Andreas Krause: conception and design of study, helped in data interpretation and evaluation of manuscript. Holger Militz: conception and design of study, supervised development of work; critically revised manuscript.

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