IMPACT STRENGTH IMPROVEMENT OF PP BASED WPC BY HYBRIDIZATION WITH PP IMPREGNATED PET FIBER

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Abstract
Polypropylene (PP) impregnated PET fiber pellets with an initial length of 15mm have been used in this study as reinforcement of PP and PP based WPC (Wood Polymer Composites).
Injection molded PP – wood flour – PET long fiber formulations showed a synergistic effect by using both fillers. The combination acts as effective impact modifier for PP and WPC.

1. Introduction

WPC (Wood Polymer Composites) are natural fiber composites (NFC) based on a polymer matrix and a wood-flour based filler, supporting a natural design. In the case of a polypropylene (PP) matrix, wood filler reinforces the stiffness of the thermoplastic matrix in injection molded parts. To further improve the impact strength performance of this material, it is either possible to improve the matrix by using a heterophasic PP copolymer or by using external added elastomers. This can lead in several cases to a significant reduction of tensile strength and stiffness.

An alternative approach is the use of polymer fibers as impact modifier for wood filled composites. From own studies it is known that short polyvinyl alcohol (PVA) fibers as filler improves the impact behavior of a PP homopolymer dramatically, and tensile properties have been improved [1]; besides, special short polyethylene terephthalate (PET) fibers improve the property profile of WPC better than elastomers [2]. In both cases, impact strength was also improved due to the pull-out process of the fibers during testing.
In the present study, we used for the first time PP impregnated long polymer fibers as impact modifier for WPC. Longer fibers should be a more effective reinforcement in thermoplastics than shorter fibers, according to results from literature on long glass fibers [3].

2. Materials and Methods

2.1. Preparation of WPC/NFC

PP types from Borealis, HJ120UB and HJ325MO as high flowability homopolymers intended for injection molding and EE050AE, have been used. HJ120UB is a special low viscosity PP homopolymer developed to fit the production of fibre reinforced thermoplastics. EE050AE is a high impact heterophasic copolymer (HECO) with an ethylene-propylene rubber content of 30 wt%.
WPC were prepared by compounding PP with wood flours from J. Rettenmaier & Söhne. Two different wood flour types, Arbocel® C320 (particle size from 100-400 µm) and Arbocel® C100 (particle size from 30-200 µm) were used. For several trials, regenerated cellulose fibrous powder (Tencel® FCP 10/300/M, from Lenzing AG) was applied. As coupling agent (CA) for the WPC or NFC compound, a maleic anhydride grafted polypropylene with high grafting level (Scona TPPP 6102 GA or Scona TPPP 8112 FA, respectively) from Byk Altana were used (5-14% based on the wood content; wood content was between 20 and 30wt%).

A parallel, co-rotating twin screw extruder Brabender DSE20 comprising a screw diameter (d) of 20 mm, and a length of 40 d was used for compounding of WPC/NFC. A twin screw sidefeeder is typically employed for feeding natural fillers. All components are fed via Motan-Colortronic gravimetric dosing scales. 2 vertical ports are used for venting. The maximum melt temperature measured at the screw tip was 187°C. For granulation, an ECON EUP50 under-water pelletizer was used.

2.2. Preparation of PP Impregnated Long Fiber Pellets (L-PET-PP)

For the presented trials, long polyethylene terephthalate (PET) fibers were chosen for impact modification. The long polymeric fibers have been impregnated and coated with PP according to the following method: an endless PET fibre was combined with the molten polypropylene base material in a continuous manner using a pultrusion process. First the PET fibre is impregnated with a first part of the polypropylene base material. Subsequently the so impregnated PET fibre is coated with the remaining second part of the polypropylene base material. The thus obtained strands of the polypropylene composition are solidified and cut into specific lengths. By this impregnation and coating with PP, long fiber PET granules were produced. The fiber length of the granules was either varied or set at 15 mm.
A PET T715 multifilament yarn (10 000 dtex) from DuraFiber Technologies was used. Coupling agent was not necessary. As seen from previous results, by using longer polymeric fibers as reinforcing filler, coupling agent is not successful for improving the impact behavior, due to the prevention of fiber pull-out.

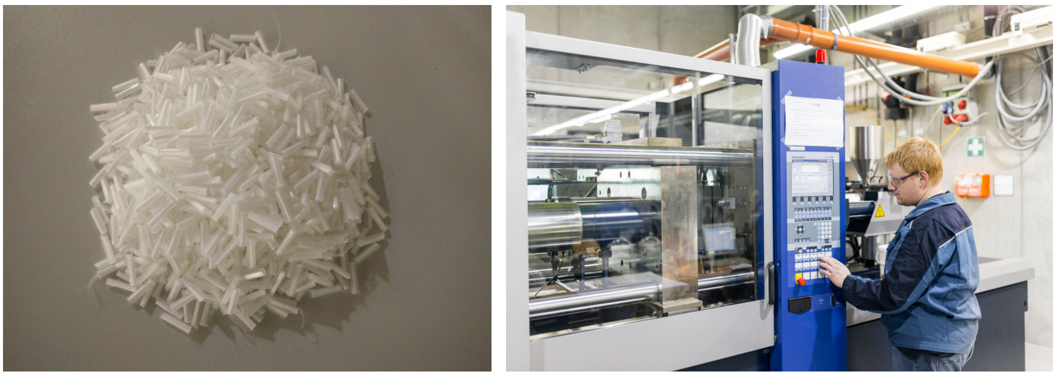


Figure 1. L-PET fiber pellets and their use in injection molding.

2.3. Injection Molding of Composites and Testing

PP impregnated long fiber PET granules (PP-L-PET) were either directly used alone or further diluted with PP granules. Before injection molding of hybrids, the long fiber PP-L-PET granules and the WPC compounds (both dried at 80°C) were cold mixed. Injection molding was then performed on a hydraulic Wittmann-Battenfeld HM 1300/350 injection molding machine, with 35 mm screw diameter and a maximum shot volume of 170 cm^3. The temperature set at the nozzle was 190°C, tool temperature was set at 60°C. Screw speed (peripheral velocity) was 250 mm/s and the back pressure was set at 50 bar. An injection speed of 50 cm^3/s was chosen for a shot volume of 45 cm^3. Packing pressure was held at 80 % of the maximum injection pressure for 15 s, while total cooling time was 24 s. Specimens with ISO 527 specified dimensions (universal testing specimen) were prepared.
The density of the specimens was determined by a buoyancy method, employing a Sartorius analytical balance, according to ISO 1183-1. Tensile properties were determined according to ISO 527; tensile strength at a loading rate of 5 mm/min or 50 mm/min, respectively. Charpy impact strength, both notched (1eA) and unnotched (1eU), were tested according to ISO 179; the heat deflection temperature was determined according to ISO 75, in mode A (HDT-A).
Residual fiber length was analyzed from injection molded specimens after PP extraction with boiling xylene, employing a Soxhlet apparatus. The isolated curved fibers, which stuck together, were then manually detached and measured by a FASEP Eco Ed. 2013 system, containing a 9000F high resolution scanner. The peak maximum of the fiber length distribution density was determined.
For determination of the final wood filler and L-PET fiber content in the hybrid composite, PP was removed by boiling in Xylene, as mentioned. The isolated long PET fibers and short wood particles, were then separated by sieving, and their fractions measured. By these calculations, the final PET fiber content and the final wood fiber content could be estimated.
Acoustic emission (AE) signals were recorded by a Sensophone AED 40/4 apparatus with a threshold value of 20 dB. A single A11 resonance detector with the resonance frequency of 150 kHz was attached to the center of the tensile specimen.
The CT scans were performed using a laboratory CT device Nanotom 180. Scanning the complete cross sectional region of a standard multi purpose test specimen lead to a voxel edge length of 5 µm. A voltage of 60 kV and Molydenum as target material were used for increased contrast. 1900 projection images and 5 times averaging per angular scanning position took 170 minutes.

3. Results and Discussion

3.1. Use of L-PET Fibers as Impact Modifier for a PP Matrix

As we see in Fig.2, if we use approximately 10wt% of L-PET fibers in the composite formulation, tensile properties will be improved and heat deflection temperature will be increased. Tensile modulus increases linearly; tensile strength, heat deflection temperature and elongation show a maximum between 20wt% and 25wt% L-PET fiber content. In particular, very high elongation values up to 26% can be achieved by using such long and ductile fibers. Fig. 3 shows that these L-PET fibers are further acting as very effective modifier in PP for improving notched impact strength.

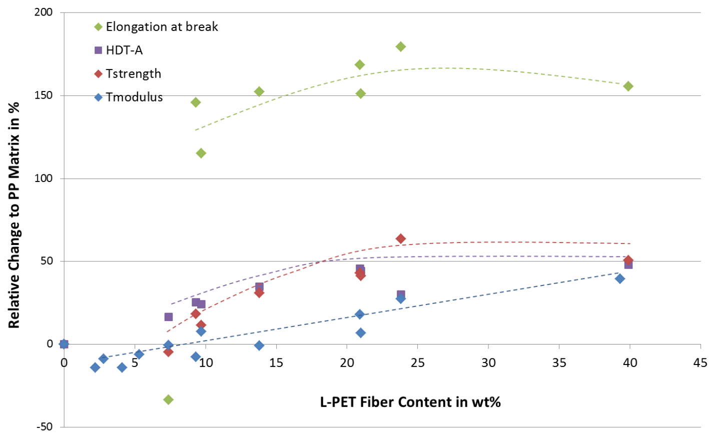


Figure 2. Relative changes of tensile properties (elongation at break, strength, modulus) and of heat deflection temperature by use of L-PET fiber pellets (15 mm) in a PP homopolymer matrix.

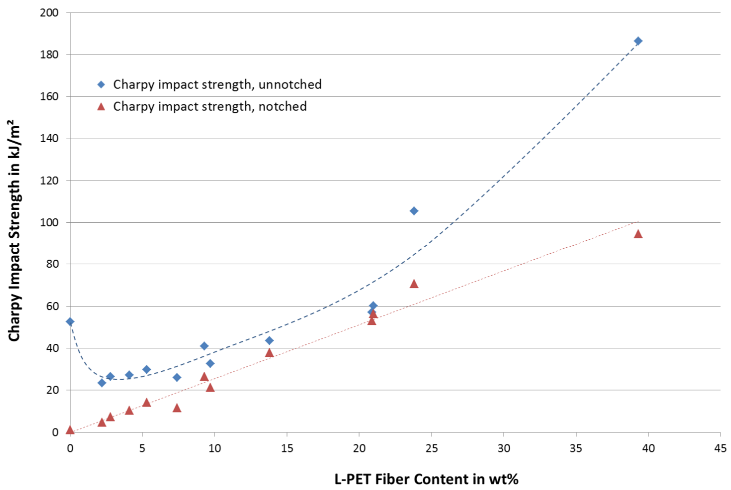
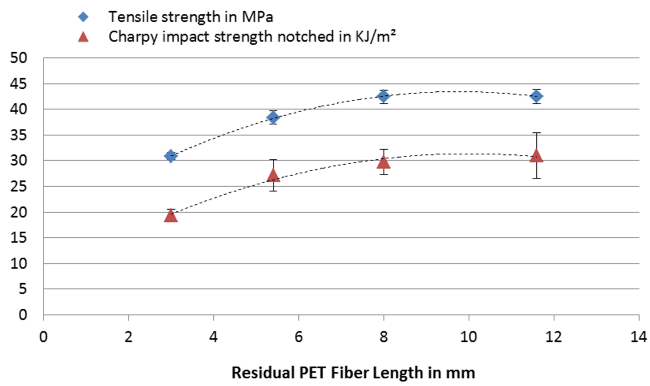


Figure 3. Changes of Charpy impact strength, notched and unnotched for the use of L-PET fiber pellets (15 mm) in a PP homopolymer matrix.

Coupling agent was not used for L-PET PP; a previous study proved that the use of maleic anhydride grafted PP results in significantly lower impact strength due to preventing of fiber pull-out. A variation of processing temperatures showed that a temperature between 180°C and 190°C at the nozzle is appropriate for injection molding of these long fiber pellets.

The increase in tensile strength and notched impact strength is depending on the fiber length used, as shown in Fig. 4a. A final fiber length of 8 mm should be achieved in the composite to obtain optimum reinforcement.
The result of acoustic emission testing is presented in Fig. 4b. A process including matrix damage, a signal with an amplitude of around 40dB, is shown.



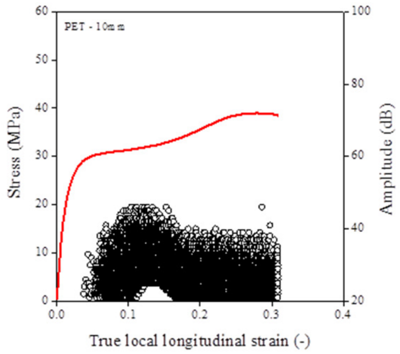


Figure 4a. Left: Tensile strength measured at 50mm/min and notched Charpy impact strength depending on the residual PET fiber length in a PP homopolymer composite (10vol% L-PET fiber). The original fiber length was 4mm, 7mm, 10mm and 15mm, respectively.
Figure 4b. Right: Result of acoustic emission testing of the 10mm L-PET PP specimen.

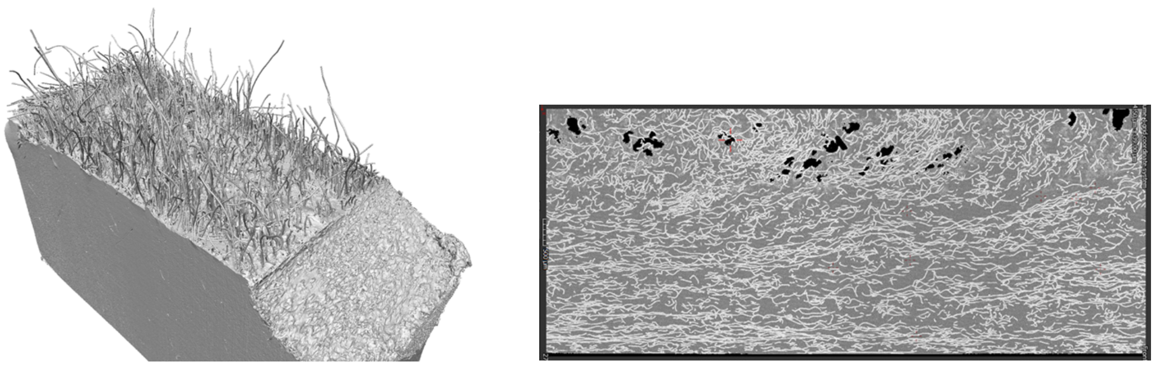


Figure 5. CT images: fracture surface after impact testing for L-PET-PP with 15mm initial fiber pellets length and fiber orientation in top view of L-PET-PP specimen with 7mm initial fiber length (both 10vol% L-PET fiber content).

The fiber pull-out behavior after impact testing and the free fiber length at the facture surface can be analyzed and presented by CT images (Fig. 5) as well as the orientation of the long curved fibers in the composite. The weighted average free fiber length for a 15mm initial fiber pellet was 800µm.

3.2. Hybrid L-PET WPC PP based Composites

The presented PP impregnated long PET fiber pellets can be further used to improve and modify PP based WPC. WPC possesses very often low impact strength and impact modification is needed. If we study hybrid fillers, it is always necessary for original lightweight composites to use an appropriate lightweight second filler. Considering the density, it is necessary to define an optimum overall filler content. For the present study low filler contents were a target.

Maleic anhydride grafted PP was used as coupling agent in the preparation of WPC, to achieve sufficient tensile properties of the WPC. However, as mentioned before, for the preparation of the L-PET PP pellets coupling agent was not used, it reduces the impact strength of both, of the L-PET PP and the hybrid composite, by preventing fiber pull-out.

In general, impact behavior of a composite is mainly defined by the used matrix material. As it is well-known, use of a heterophasic PP copolymer can distinctly improve the impact strength compared to a PP homopolymer matrix.

If we start applying a PP based WPC compound with 16wt% Arbocel C100 for injection molding, we get the following properties, see Table 1 (formulation A). By a 50/50 weight percent mixture of a PP-WPC (approx. 25 wt% Arbocel C100) with L-PET PP pellets (approx. 10 wt% PET fiber content), we prepared a hybrid composite by injection molding with finally 11 wt% wood and 6 wt% L-PET fiber content (formulation B). With similar density as before, we indeed got analogous tensile properties, but much higher notched impact strength (8 times increased). By changing the WPC formulation to a heterophasic copolymer matrix we can obtain hybrid composites with even better impact behavior and especially higher unnotched impact strength (formulation C). These changes and improvements are further illustrated in Fig. 6. Wood flour is needed as reinforcement in this formulation to achieve sufficient stiffness with tensile moduli over 2 GPa.

Table 1. Properties of PP-based WPC and of selected L-PET WPC PP hybrid composites (density; tensile modulus; tensile strength, elongation at Fmax tested with 5mm/min; Charpy impact strength unnotched 1eU and notched 1eA).

Composite Formulation	*Density* (gcm^{-3})	E_T (GPa)	σ_{max} (MPa)	ε_{max} (%)	Charpy Impact Strength (kJ/m²)	
					1eU	1eA
A:PP wood (16wt% Arbocel C100)	0.96	2.3	32	3.6	17	1.5
B: PP – L-PET wood (11 wt% Arbocel C100 + 6 wt% L-PET)	0.97	2.3	34	4.3	21	12
C: PP copolymer – L-PET wood (11 wt% Arbocel C320 + 7 wt% L-PET)	0.97	2.1	30	5.8	31	19

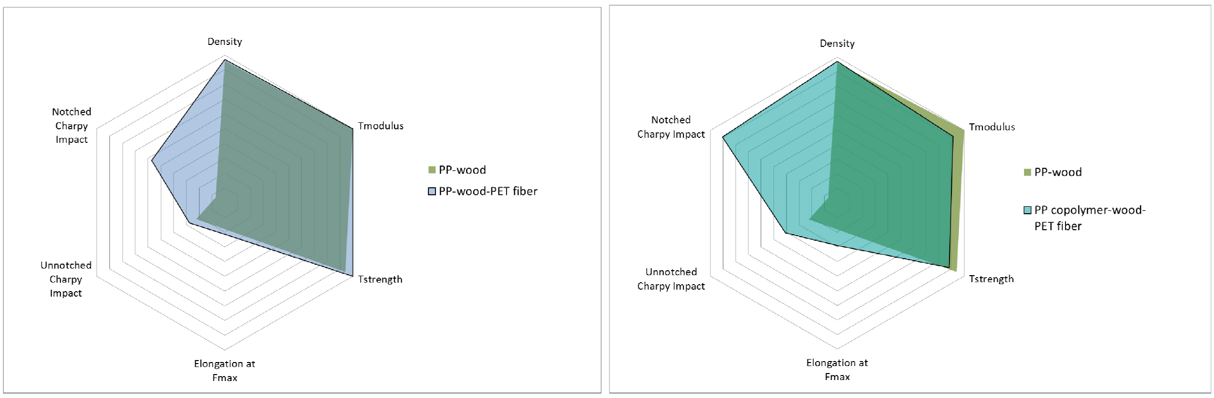


Figure 6. Illustrated comparison of formulation A (PP-wood) with B (PP-wood-PET-fiber) left and comparison of formulation A with formulation C (PP copolymer-wood-PET fiber hybrid) right.

The tensile properties, stiffness and tensile strength of such hybrid PP-L-PET wood composites can be further adjusted by the wood content and the content of copolymer matrix, as illustrated in Fig. 7.

Higher wood content and lower copolymer content in the PP matrix increase stiffness and tensile strength.

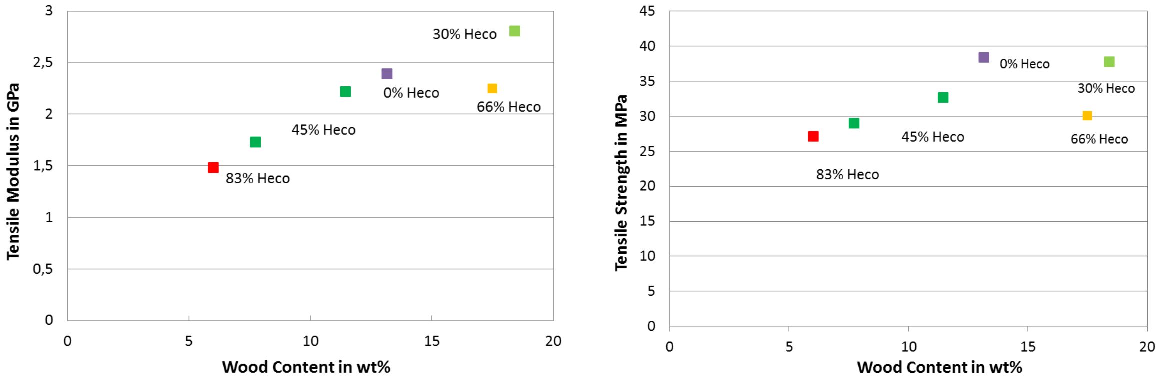


Figure 7. Tensile modulus and tensile strength for PP L-PET wood composites in dependence of the wood content (Arbocel C320) in the composite and of the content of PP heterohpasic copolymer (Heco) related to the PP matrix system. Target L-PET content was 5wt%.

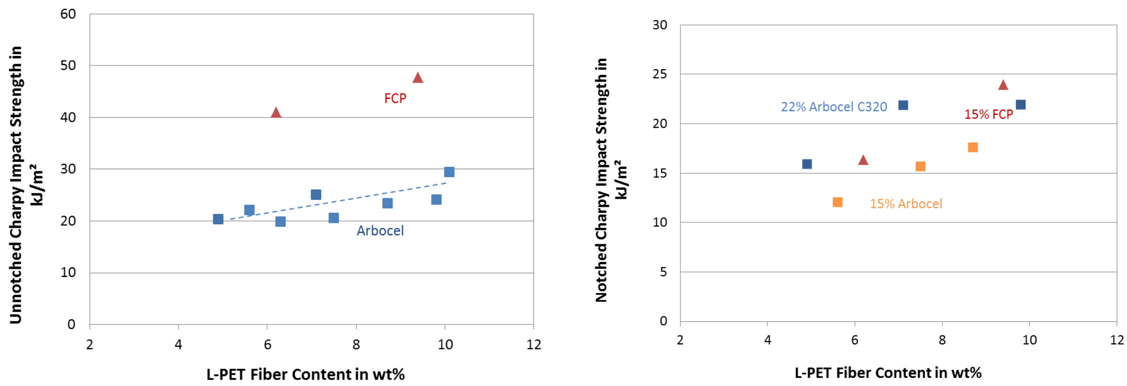


Figure 8. Unnotched and notched Charpy impact strength of PP homopolymer L-PET fiber and cellulose hybrid composites depending on the L-PET fiber content.

In Figure 8 the dependence of the impact behavior of PP homopolymer L-PET fiber and cellulose hybrid composites is presented. Unnotched impact strength depends mainly on the L-PET content and on the type of wood or cellulose used (Arbocel vs. cellulose fibrous powder FCP). The content of the cellulosic filler was not decisive. Notched impact strength of the hybrid composites depends on the L-PET content as well as on the wood content and on the type of cellulosic filler used.

4. Conclusions

A hybrid filler combination that consists of more than one filler type, can use the advantages of both fillers. In the optimum case, hybrid fillers act synergistically to each other. The studied PP composites filled with long PET fibers and wood particles combine elastic behavior and a reinforcing effect by the use of wood particles. Besides, both filler types can be used for low-weight materials.
In the present study it was shown, that addition of wood flour helps to improve tensile properties and addition of long PET fibers increases the notched impact strength significantly in comparison to a PP homopolymer matrix composite. By the use of a PP heterophasic copolymer matrix the impact

behavior can be further improved. The initial L-PET fiber length was a decisive factor to achieve a good performance in PP.

Acknowledgments

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