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The influence of mechanical recycling on properties in injection molding of fiber-reinforced polypropylene

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ABSTRACT

Due to higher mechanical demands on technical parts, the application of short fiber reinforced thermoplastics for injection molding is strongly increasing. Therefore, more attention needs to be paid to the optimization of their recycling processes. Mechanical shredding of thermoplastics into granules is a common recycling method within polymer industries. The breaking of polymer chains and reinforcing fibers during this process may affect the material properties. This study presents the effect of ten recycling sequences on four different materials: polypropylene, glass fiber filled polypropylene, carbon fiber filled polypropylene and flax fiber filled polypropylene. Tests indicate that recycling has a negative influence on most of the mechanical properties. Polypropylene without fibers forms an exception as it does not exhibit any significant change in material properties. Glass fiber and carbon fiber reinforced polypropylene show a decrease in stiffness and tensile strength during the recycling steps. The impact strength of carbon and flax fiber reinforced polypropylene increases whereas that of glass fiber reinforced polypropylene decreases.

INTRODUCTION

Since thermoplastic polymers are lightweight, inexpensive and easy to process, they have replaced traditional materials such as wood, ceramics and metals in various applications. The introduction of fiber reinforced polymers, which show an outstanding stiffness to density ratio, has even further boosted this

evolution. With a production of 1.118 million tons of glass fiber reinforced plastics in 2017 (accounting for approximately 95 % of the total fiber reinforcements within polymers) the production of fiber-reinforced plastics in Europe is already immense and is only expected to grow (annual growth of approximately 2 %) (Witten et al., 2017). Injection molding has the world's highest market share in processing these composites (Gutiérrez et al., 2013; Witten et al., 2017). However, the biggest challenge of fiber-reinforced composites is their end-of-life difficulties (Oliveux et al., 2015). Specifically, waste disposal in landfills is becoming restrictive due to its low sustainability, increasing cost, decreasing available space and restrictions by the European Union's Waste Framework Directive (Jacob, 2015).

The first major focus on large scale recycling of plastics occurred in the mid-seventies, due to a shortage of resin caused by the oil embargo and scarce manufacturing capacities (Akovali et al., 1998). While during these years most of the research focused on the recycling of non-reinforced polymers (Driscoll, 1977; Leidner, 1981; Shea et al., 1975), some authors evaluated the effect of recycling on the properties of fiber reinforced composites (Abbas, 1980; Filbert, 1968; Yang et al., 1979). In the following years, theoretical algorithms were developed to predict the resulting properties of the recycled polymers. A first theory was presented by Abbas et al. in 1978 (Abbas et al. 1978). An alternative methodology for the determination of the final product properties was proposed by Throne using a single-pass property (Throne, 1987). Bernardo, Cunha and Oliveira developed an algorithm to predict a great variety of important properties of mixtures of virgin and recycled polymers (Bernardo et al., 1993).

Until now, many different recycling techniques have been studied such as: mechanical recycling, pyrolysis, and solvolysis (Oliveux et al., 2015). Mechanical recycling is the most popular recycling method, accounting for 5 million tons of recycled material each year (Doubai et al., 2015). This recycling technique consists of grinding or shredding the polymer material into smaller pieces which allows to reuse the material in an injection molding process.

During mechanical recycling of fiber-reinforced polymers, both the molecular chains of the matrix material and the reinforcing fibers break which causes multiple disadvantages to the quality of the resulting product: quality reduction of the surface appearance, a change in thermal properties and deterioration of physical and mechanical properties, etc. (Aksesson et al., 2015; Bajracharya et al., 2014; Doubai et al., 2015; Ignatyev et al., 2014; La Mantia, 2003; Mehat and Kamaruddin, 2011). Since fiber-reinforced products are usually used for applications with high demands on mechanical properties, these properties after recycling are generally the primary concern. Several studies have determined the influence of mechanical recycling on the resulting mechanical properties of injection molded glass fiber

reinforced materials. These studies have shown that the tensile modulus, tensile strength, flexural modulus and flexural strength of the recycled material decrease compared to the virgin material. In contrast, the tensile strain at break and impact strength of the recycled material increase (Bernasconi et al., 2007; Bourmaud and Baley, 2007; Colucci et al., 2017; Giraldi et al., 2004; Kuram et al., 2014; Kuram et al., 2016). During all these experiments, a maximum of five recycling steps were executed. However, results indicate that the effect of recycling on the mechanical properties was not stabilized yet. Other studies determined the influence of mechanical recycling of flax fiber reinforced polymers, but these results are often inconsistent. Le Duigou et al. found that the tensile modulus, tensile strength and strain at break all decreased over six recycling steps for poly(L-lactide) containing 20 % and 30 % flax fibers by weight. The tensile modulus is only slightly influenced by recycling as it only decreased by 10 %. The tensile strength was halved over six recycling steps and the strain at break had a reduction of 65 %. Gourier et al. studied the recycling stability of polyamide 11 and polypropylene, both reinforced with 50 vol% flax fibers. The authors indicate that for the polypropylene composite, the tensile modulus, tensile strength and strain at break remained relatively stable throughout six recycling steps. However, for the polyamide composite, the tensile modulus and tensile strength decreased. In contrast, the strain at break increased to 300 %. Unfortunately, both of the studies on flax fibers did not look into the effect of recycling on the impact strength.

The aim of the present study is to further investigate the effect of mechanical recycling on the mechanical properties of injection molded fiber-reinforced polymers. The flexural modulus, tensile strength, strain at break and impact strength of different fiber reinforced polymers are measured. For this study, polypropylene with three different types of fibers are used: glass fibers, carbon fibers and flax fibers. A fiber concentration of 20 wt% was used for each material. To the author's knowledge, no literature could be found on the effect of mechanical recycling on properties of injection molded carbon fiber reinforced polymers. Since in current studies, the number of recycling steps were limited, this study uses ten consecutive injection molding and recycling steps. To eliminate the potential effect of recycling on the mechanical properties of the polypropylene material itself, comparative tests were also carried out for non-reinforced polypropylene.

MATERIALS AND METHODS

Materials

For this study, four grades of polypropylene were selected: (1) A general purpose polypropylene (PP) grade PHC31 from Sabic. (2) A 20 wt% glass fiber filled polypropylene (PP GF) grade G3220A from Sabic which is commonly used in the automotive industry. (3) A 20 wt% carbon fiber filled polypropylene (PP CF) grade Stat-Tech PP20CF from PolyOne. The carbon fibers in this material add reinforcing properties as well as conductive properties. (4) A 20 wt% natural (flax) fiber filled polypropylene (PP NF) grade 036CG from Beologic. Mechanical properties of these four materials as provided by suppliers are listed in Table 1.

TABLE 1 Mechanical properties of the different materials provided by suppliers (* Notched impact strength).

	PP	PP 20% GF	PP 20% CF	PP 20% NF
Tensile strength (MPa)	25	80	45	28
Flexural modulus (MPa)	1300	4000	9500	1950
Impact strength (kJ/m ²)	12.5*	46	12	20
Fiber content (wt%)	0	20	20	20

Methods

Sample production

Samples were produced on an Engel ES 200/35 HL hydraulic injection molding machine with a maximum clamping force of 350 kN and a 25 mm screw with an L/D ratio of 24.8. The mold temperature is controlled by a Wittmann Temprow D controller.

The injection molding parameters used in this study are listed in Table 2. The injection temperature and mold temperature were set as recommended by the material supplier. The volumetric injection rate, back pressure and screw rotation speed was set to a low value in order to reduce fiber breakage due to shear stress. The implemented holding pressure was an intermediate value between an upper limit corresponding to flash and a lower limit corresponding to sink marks.

TABLE 2 Process parameters for the different materials.

	PP	PP 20% GF	PP 20% CF	PP 20% NF
Injection temperature (°C)	230	230	230	190
Volumetric injection rate (cm ³ /s)	39	39	39	39
Back pressure (bar)	80	80	80	80
Screw rotation speed (m/s)	0.25	0.25	0.25	0.25
Holding pressure (bar)	220	475	285	555
Mold temperature (°C)	30	40	70	30

The injection molded specimen selected for this study consists of two ISO 527-1/1B tensile bars with a thickness of 4 mm, as shown in Figure 1.

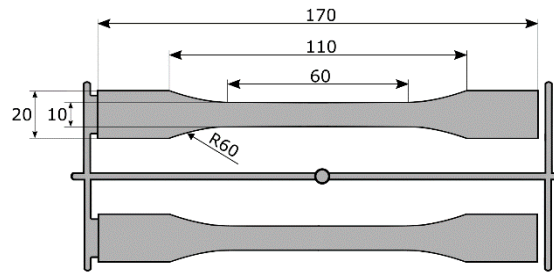


FIGURE 1 Geometry of the ISO 527-1/1B samples with dimensions. The thickness of the part is 4 mm.

Recycling process

The recycling process is illustrated in Figure 2. During the first sequence, virgin granulate was molded into tensile bars. Next, these samples were shredded to obtain grinded polymer which was then used for injection molding of new samples. No additional compounding step was done in between. The shredder used for the recycling process was a Rapid Granulator 1521 which grinds the products into granules from 2 to 6 mm. The recycling sequence was repeated ten times for each material and started from the previously grinded polymer. As a consequence, the process did not deal with post-consumer recycling of soiled, contaminated materials. During each cycle, thirty samples were collected for evaluation of the mechanical properties.

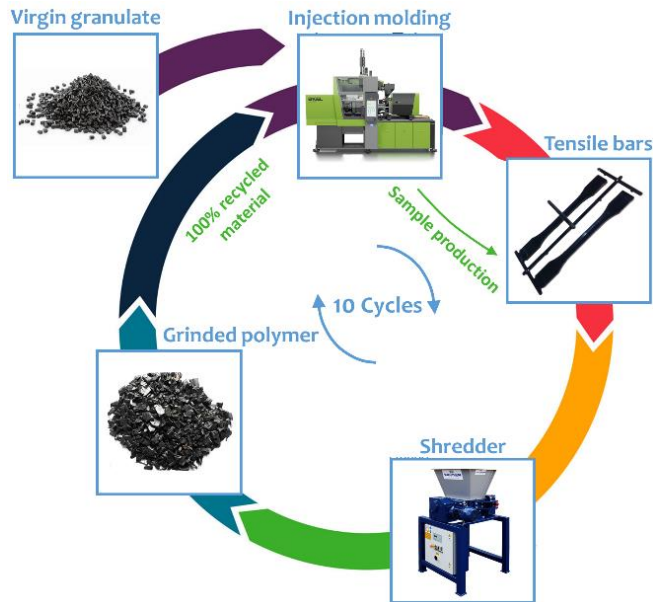


FIGURE 2 Schematic representation of the recycling process.

Tensile tests

Tensile tests were executed on five samples during each evaluation to determine the maximum tensile strength of the material. The average tensile strength along with its 95 % confidence interval is reported for each setting. The tensile tests were performed according to ISO 527-1:1993, at a speed of 100 mm/min, on a Galdabini Quasar 50 with a 50 kN load cell.

Three point bending tests

Three point bending tests were carried out on five samples during each evaluation to determine the flexural modulus. The average flexural modulus along with its 95 % confidence interval is reported for each setting. The bending tests were performed according to ISO 178:1993, at a speed of 2 mm/min, on a Zwick BZ2.5/TS1S 3 with a 500 N load cell.

Impact tests

Impact tests were performed according to Charpy ISO 179:1993, on a Zwick 5113 pendulum impact tester with an 7.5 J impact hammer. For the non-reinforced PP samples, a notch according to ISO 179/1_e_A was used as the unnotched samples did not break. The reported impact strength values represent the average out of five different samples along with a 95 % confidence interval.

Analysis of fiber length distributions

An indication on the fiber length distribution was obtained by measuring the length of fibers extracted from injection molded samples. Samples containing carbon and glass fibers were placed in an oven at 450 °C, causing the sublimation of the polymer material and hence the extraction of the fibers. Next, these fibers were dispersed on a glass microscope slide and visualized using a digital microscope. This technique could not be used for the flax fiber reinforced parts as these fibers would also sublimate. However, as the polymer is translucent, embedded fibers at the surface of these parts could be observed. The microscope used in this study is a Keyence VH-S30 digital microscope with a maximum magnification of 200 connected to a VHX-500F monitor with build-in measuring software. Each fiber was manually selected and defined with a two point measuring bar. For each sample, about 100 fibers were examined and statistically analyzed to give an indication on the average fiber length distribution. Fibers shorter than 35 μm and fibers intersecting with the edges of the picture were not taken into account.

RESULTS AND DISCUSSION

Tensile strength

Figure 3 shows the tensile strength (a) and the tensile strength relative to the first sequence (b), both in function of the recycling steps for each of the materials. PP GF has the highest initial tensile strength (76 MPa) followed by PP CF (47 MPa). According to literature, glass fibers have a higher tensile strength than carbon fibers which explains these values (Ku and Wang, 2011). The tensile strength of PP NF is only slightly higher than that of non-reinforced PP. The tensile strength for both materials remains almost constant during the recycling sequences. In contrast, the tensile strength of PP GF and PP CF show a non-linear decreasing trend with a reduction in tensile strength of respectively 50 % and 25 % after ten recycling steps. The decline is the largest at the beginning of the recycling steps and stagnates towards the end. Although PP GF shows the largest decrease, after ten recycling steps it still is the material with the highest tensile strength.

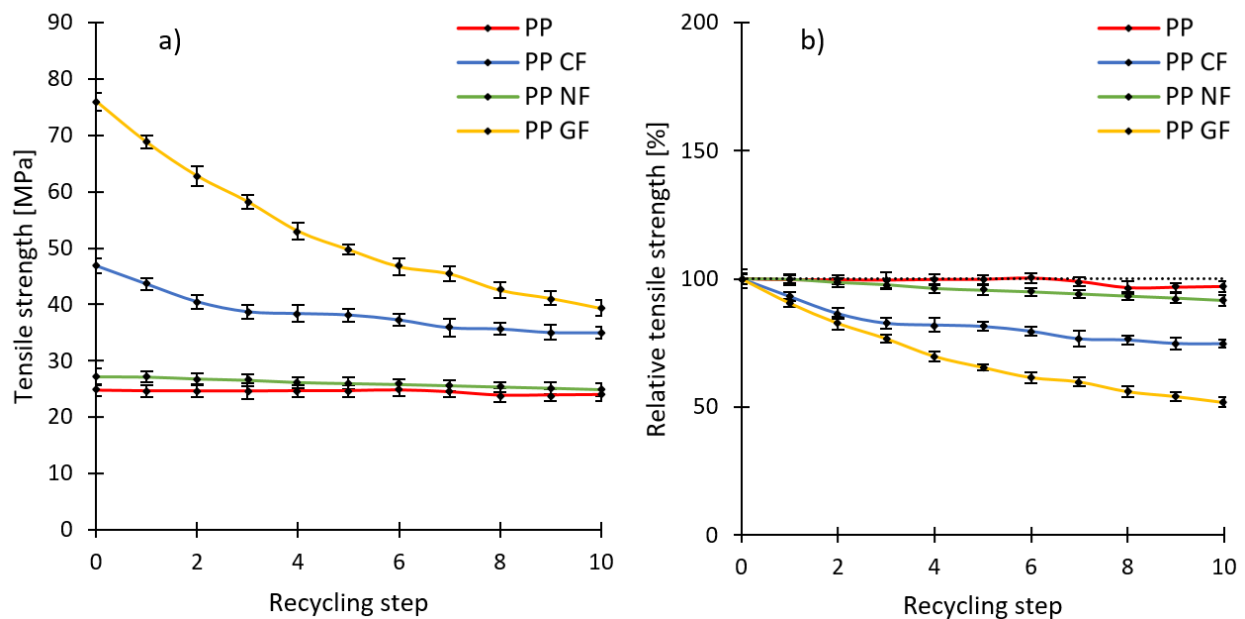


FIGURE 3 Representation of the tensile strength in function of the recycling sequence for each of the materials absolute tensile strength (a), tensile strength relative to the first sequence (b). The error bars represent the 95 % confidence interval.

Flexural modulus

Figure 4 shows the absolute flexural modulus (a) and the flexural modulus relative to the first sequence (b), both in function of the recycling sequences for each of the materials. PP CF has the highest initial flexural modulus (9320 MPa), followed by PP GF (4000 MPa). This trend can be attributed to the higher stiffness of carbon fibers compared to glass fibers, as stated in literature (Ku and Wang, 2011). The flexural modulus of PP NF shows only a minor increase compared to PP. Furthermore, after ten recycling steps, the flexural modulus of both PP NF and PP is reduced by 10%. In contrast, the flexural modulus of PP GF and PP CF show a decreasing trend of respectively 45 % and 35 %. Similar to what was seen for the tensile strength, PP CF retains the highest value for the flexural modulus after ten recycling sequences.

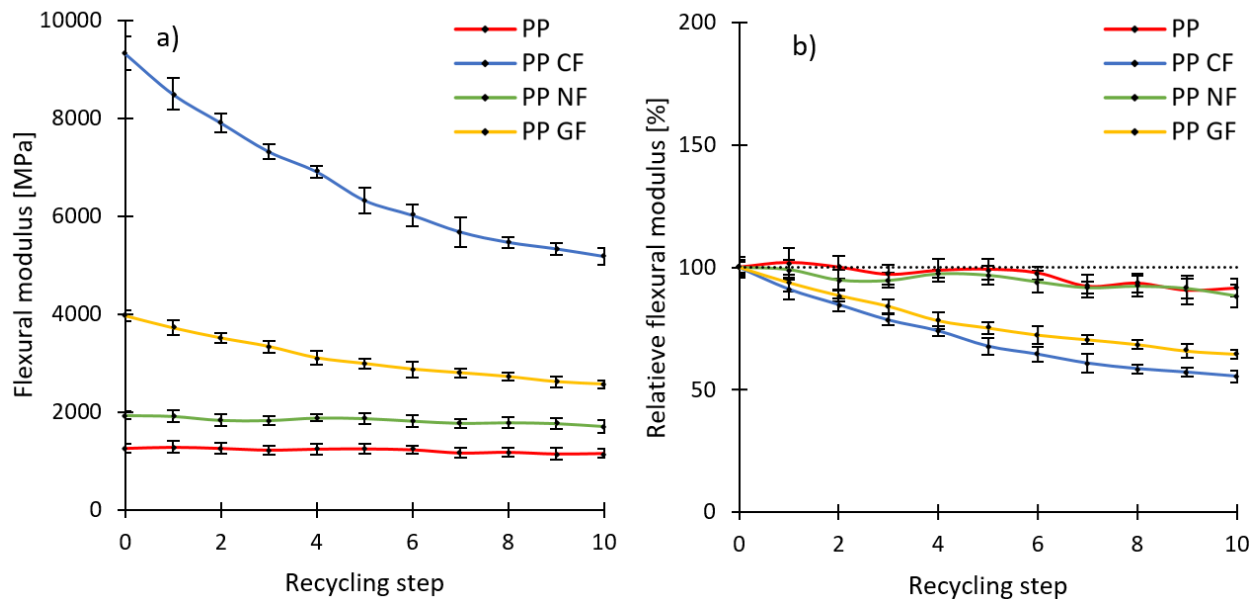


FIGURE 4 Representation of the flexural modulus in function of the recycling sequence for each of the materials absolute flexural modulus (a), flexural modulus relative to the first sequence (b). The error bars represent the 95 % confidence interval.

Impact strength

Figure 5 shows the absolute impact strength (a) and the impact strength relative to the first sequence (b), both in function of the recycling sequences for each of the materials. The non-reinforced PP samples did not break during tests and are therefore not represented in the figure. The fiber-reinforced material

with the highest initial impact strength is PP GF (42 kJ/m²) followed by PP NF (19 kJ/m²), while PP CF shows the lowest initial impact strength (12 kJ/m²). It is remarkable that the impact strength of PP GF decreases in function of the number of recycling steps (approximately 50 %), while the impact strength of PP CF and PP NF increases (approximately 50 %).

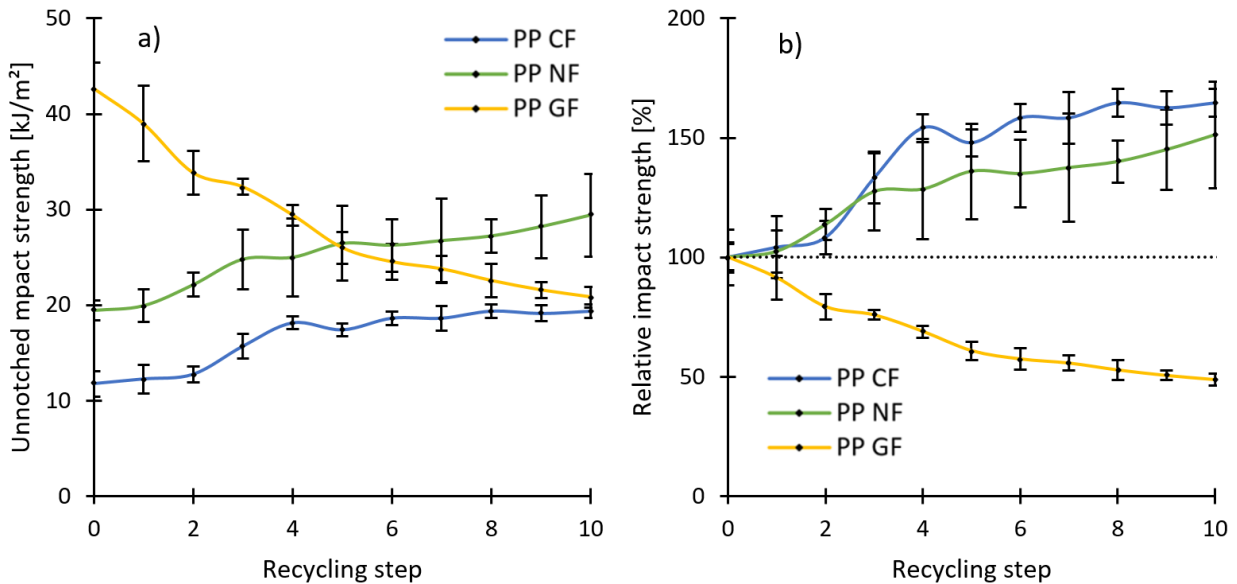


FIGURE 5 Representation of the impact strength in function of the recycling sequence for each of the materials absolute impact strength (a), impact strength relative to the first sequence (b). The error bars represent the 95 % confidence interval.

Stress-strain curve

Figure 6 illustrates a stress-strain curve for each recycling sequence of each material. The intensity of the color for each of the materials diminishes as the number of recycling sequences increases. The moment of rupture is indicated by a square symbol for PP GF, a triangle symbol for PP CF and a circular symbol for PP NF. The graph summarizes all previously discussed material properties. The highest value for the stress indicates the tensile strength, while the slope of the linear area in the beginning of the curves is an indication of the stiffness.

PP CF shows the highest stiffness followed by PP GF, PP NF and PP. Throughout the recycling steps, a significant decrease in stiffness can be observed for PP CF and PP GF. PP without fibers and PP NF did not experience any significant change in stiffness. PP GF has the highest tensile strength which decreases considerably during the recycling steps. What stands out is that the elongation at break

remains almost constant for PP GF, while it increases noticeably for PP CF and PP NF. The highest strain at break is observed for PP, for which the material breaks at $\pm 75\%$ strain throughout all the recycling steps. Moreover, curves of non-reinforced PP all have a similar shape and are thus layered on top of each other.

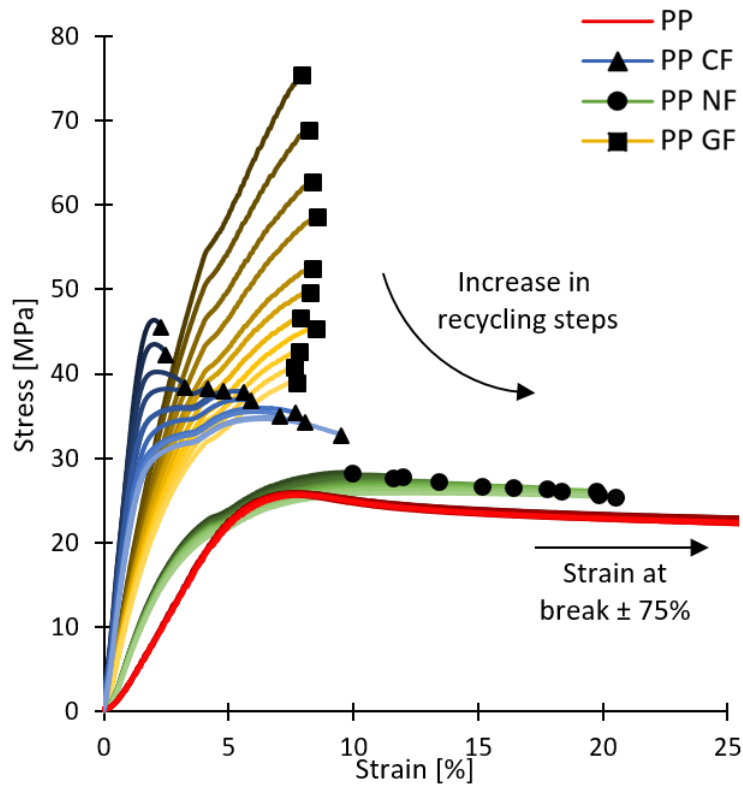


FIGURE 6 Representation of the stress strain curve in function of the recycling sequence for each of the materials.

Effect of recycling on the fiber length distribution

Figure 7 shows a microscope image with related fiber length distribution for carbon fibers after one (a), five (b) and ten (c) injection molding cycles. Smaller fibers can already be observed after one injection cycle. As these fibers are not recycled yet, it can be assumed that the smaller pieces are a result of fiber length reduction during the injection molding process. However, fiber lengths larger than 600 μm were still obtained. After five consecutive recycling and injection molding steps, a significant reduction in fiber length can be observed. The amount of smaller fibers has increased and the amount of large fibers has decreased. This reduction could be attributed to both the fiber breakage during injection molding and the mechanical recycling process. The influence of these individual processes is not determined in this study. However, according to the Colucci et al. (2017), the reduction is mainly caused by the mechanical recycling process. After ten consecutive recycling and injection molding steps, the fiber length reduction has even further increased as there is a very high frequency (> 75 %) of small fibers (< 100 μm).

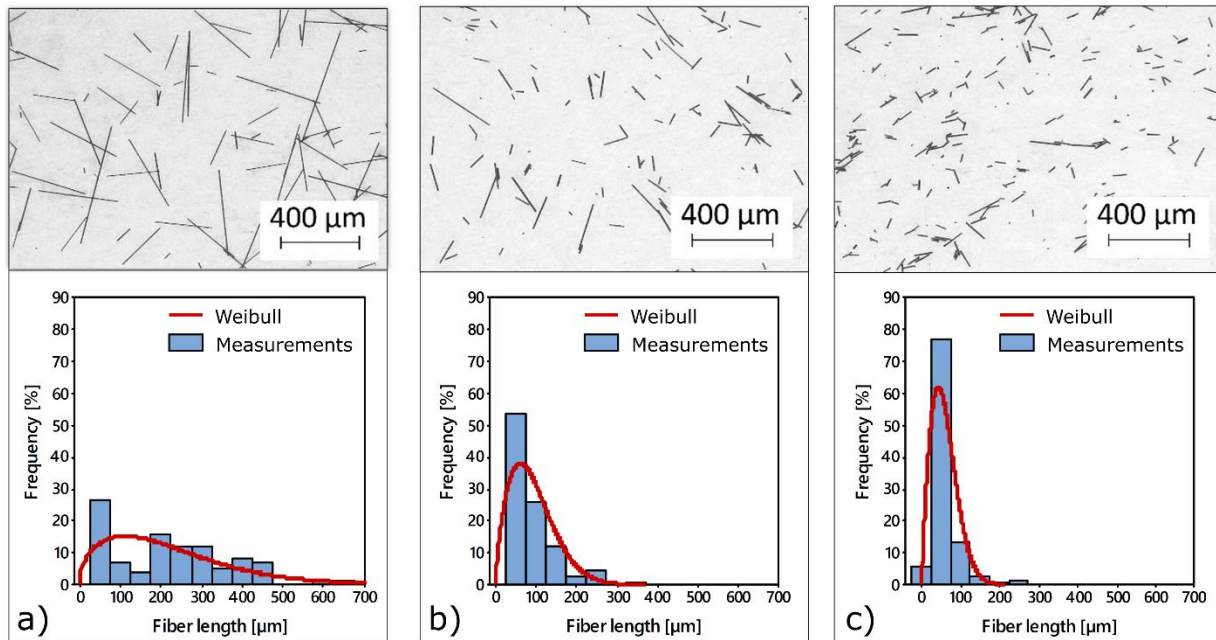


FIGURE 7 Microscopic image with related fiber length distribution for carbon fibers after one (a), five (b) and ten (c) injection molding cycles; corresponding Weibull distributions are also plotted.

Figure 8 shows a microscope image with related fiber length distribution for glass fibers after one (a), five (b) and ten (c) injection molding cycles. Like for the carbon fibers, also for the glass fibers a number of low length fibers can be observed after one injection molding cycle. However, the largest glass fibers show a length up to 1800 μm , which is significantly higher compared to carbon fibers. As the amount of recycling steps increases, the distribution of the fibers decreases, resulting in a large amount of small fibers. After ten consecutive recycling and injection molding steps all examined fibers show a length smaller than 600 μm .

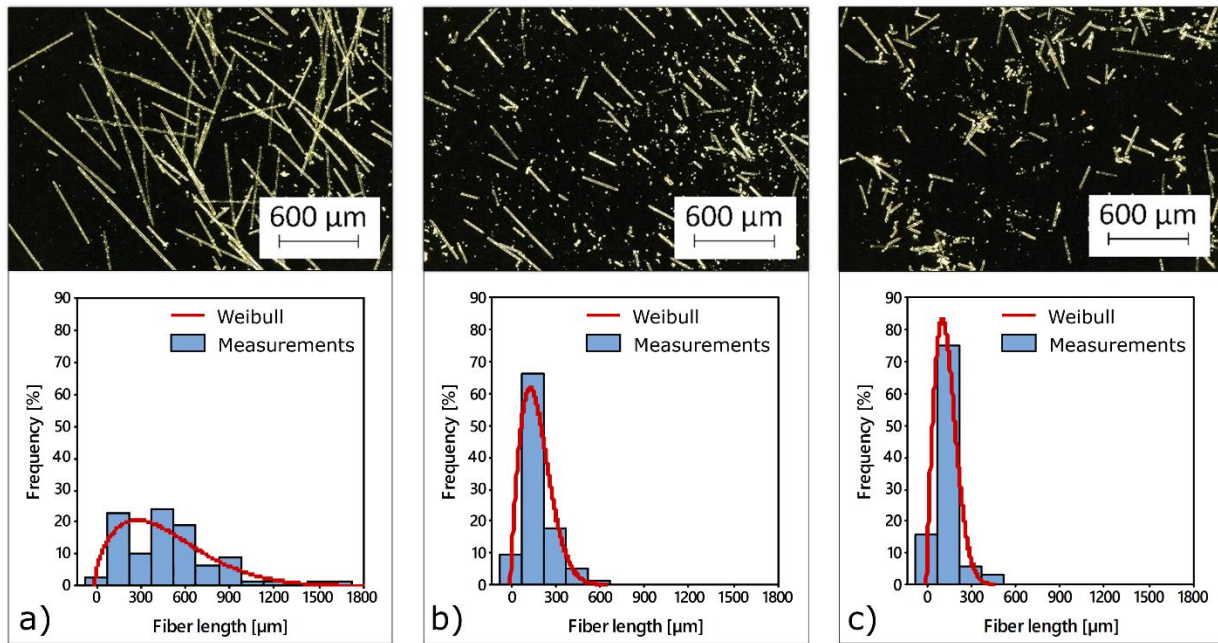


FIGURE 8 Microscopic image with related fiber length distribution of glass fibers after one (a), five (b) and ten (c) injection molding cycles; corresponding Weibull distributions are also plotted.

Figure 9 shows a microscope image of flax stems with related stem length distribution after one (a), five (b) and ten (c) injection molding cycles. The examined flax stems consist of three layers: an inner core of xylem, a layer of flax fiber bundles and an outer layer of bark (Zhu et al., 2013). The initial stem length and width are drastically larger compared to glass and carbon fibers. Therefore, only a slight amount of small stems are observed after one injection molding step. As the recycling steps increase, both width and length of the stems decrease.

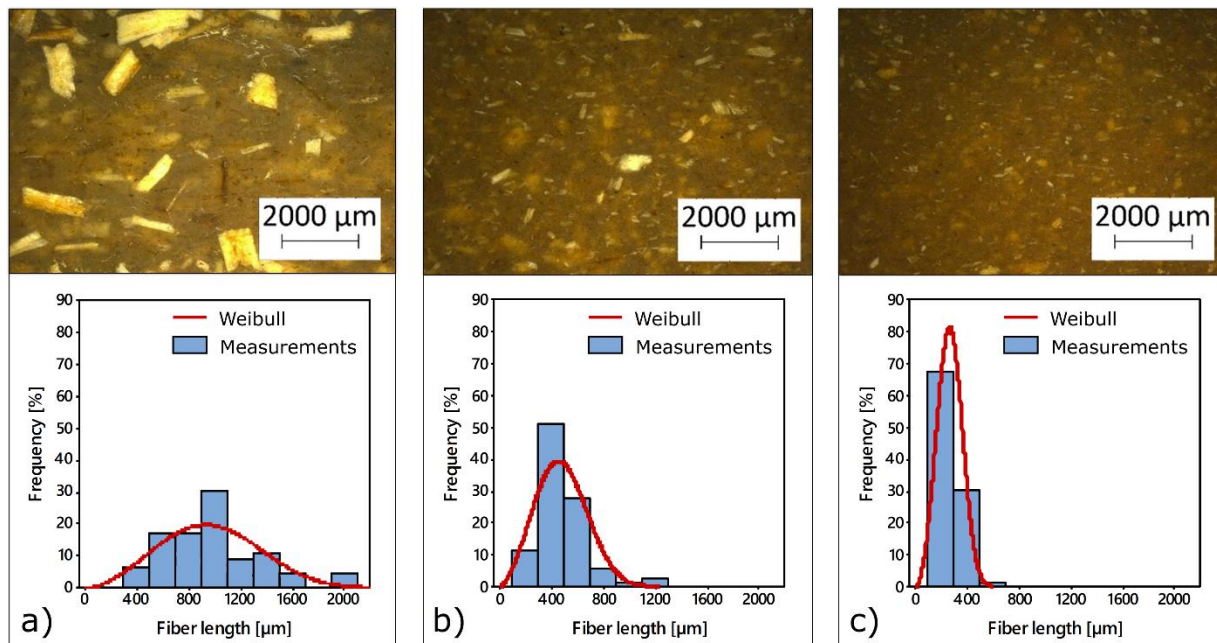


FIGURE 9 Microscopic image of flax stems with related stem length distribution after one (a), five (b) and ten (c) injection molding cycles; corresponding Weibull distributions are also plotted.

Discussion

The addition of fibers in polypropylene results in an increased flexural modulus and tensile strength. In contrast, the strain at break and impact strength are decreased. Carbon fiber reinforced polypropylene shows the highest increase in stiffness compared to non-reinforced polypropylene (740 %), whereas an addition of glass fibers lead to the highest increase in tensile strength (300 %). Natural fibers only provide a slight increase in tensile strength (112 %) and flexural modulus (153 %).

The mechanical properties of non-reinforced polypropylene remain almost constant throughout all of the mechanical recycling steps. The relative decay in properties after ten recycling steps is < 5 %. Hence, in this work it can be assumed that the mechanical properties of the polypropylene material itself are not affected by recycling.

Mechanical recycling of fiber-reinforced polymers results in a reduction of the tensile strength and flexural stiffness for all of the observed composites. The composites with carbon and glass fibers show a considerable reduction throughout ten recycling steps. In contrast, the flax fiber filled material shows only a slight decrease. The materials with the highest initial properties show the highest reduction. The declining trend in tensile strength and bending stiffness in PP CF and PP GF is exponential and stagnates towards ten recycling steps. This trend is caused by the fiber breakage, induced by the mechanical recycling. This is in agreement with Thomason's (2002) statement that smaller fibers have less influence on the mechanical properties compared to larger fibers. The initial fibers have a high length and therefore break more easily. Towards the end of the recycling steps, the fibers are much smaller and less likely to break. As a result, after ten recycling steps the reduction in fiber length stabilizes, thereby stopping the reduction of the mechanical properties. From an industrial point of view, it is recommended to add a concentration of virgin material to the recycle after each recycling sequence, as this will minimize the reduction in mechanical properties of the material.

According to this study, the mechanical recycling of fiber-reinforced polymers results in an increase of strain at break and impact strength for carbon and flax fiber filled polypropylene. In contrast, the strain at break and impact strength of glass fiber reinforced polypropylene show a large reduction throughout ten recycling steps. This contradictory result could be attributed to the critical fiber length, which is defined as the fiber length required for the fiber to develop its fully stressed condition in the matrix. If the fiber is shorter than this critical length, it will easily slip from the matrix (fiber pull-out failure),

inducing an increase in elongation at break (Bourmaud and Baley, 2007; Kuram et al., 2016). The critical fiber length can be determined using the Kelly-Tyson equation (Kelly and Tyson, 1965):

$$l_c = \frac{\sigma_f \cdot d}{2 \tau_c} \quad (1)$$

where σ_f is the fiber longitudinal tensile strength, d is the fiber diameter and τ_c is the maximum interfacial shear stress. When comparing these parameters for the studied fibers, a large difference can be observed in the interfacial shear stress. Glass fiber reinforced polypropylene has a high interfacial shear stress (e.g. $\tau_c = 31.8$ MPa) indicating a high compatibility and a low critical fiber length (e.g. $l_c = 0.4$ mm) (Yan and Cao, 2018). Carbon fiber and flax fiber reinforced polypropylene have a very low interfacial shear stress (e.g. $\tau_c = 3.6$ MPa and $\tau_c = 12.0$ MPa resp.) indicating a low compatibility and a high critical fiber length (e.g. $l_c = 3.6$ mm and $l_c = 3.2$ mm resp.) (Huber and Müssig, 2008; Yan and Cao, 2018). To improve the poor compatibility, several chemical modifications or fiber sizing are applied to improve the interfacial matrix-fiber bonding (Bourmaud and Baley, 2007; Ku and Wang, 2011; Pickering et al., 2016). It is shown that the fiber length decreases after mechanical recycling. Moreover, it could be possible that mechanical recycling also affects fiber sizing, leading to a decreased compatibility between fiber and matrix material. The resulting fiber length of the recycled carbon and flax fiber composites will be much smaller than the critical fiber length and the fibers will slip from the matrix, thus causing an increased strain at break. As a consequence, the area under the stress-strain curve, which correlates to absorbed energy by the material, is also increased. Thus, the increased strain at break might also be an explanation for the increased impact strength, observed for the carbon and natural fiber reinforced polypropylene. In contrast, the glass fiber reinforced composite has a small critical fiber length, thus resulting in a constant strain at break.

After tensile tests were conducted, a striking difference in fracture surface could be noticed between the different materials. The carbon fiber and flax fiber reinforced specimens showed an irregularly shaped fracture with fibers sticking out of the ruptured plane as presented in Figure 10 (a). This could confirm that the fiber length is smaller than the critical fiber length, resulting in a fiber pull-out failure mechanism. Figure 10 (b) shows the fracture of glass fiber filled specimens. A straight fracture without noticeable fibers is observed which indicates the fiber fracture failure mechanism (Cantwell and Morton, 1991).

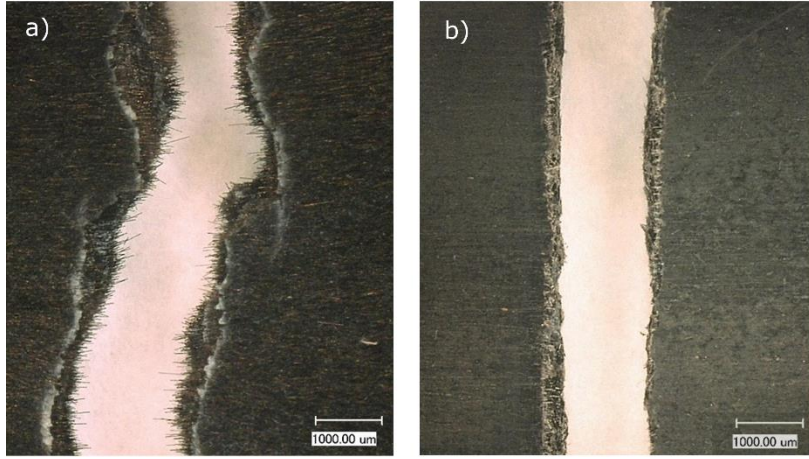


FIGURE 10 Microscopy photo of the ruptured sample after tensile tests for carbon fibers (a) and glass fibers (b).

CONCLUSIONS

In this study, the effect of mechanical recycling on the mechanical properties of injection molded fiber-reinforced polypropylene was investigated. Mechanical recycling of fiber-reinforced polymers caused a reduction of the tensile strength and flexural stiffness for all studied composites. The highest reduction in tensile strength was observed for the glass fiber reinforced polypropylene while carbon fiber reinforced polypropylene showed the highest reduction in flexural stiffness. The flax fiber filled polypropylene only showed a slight decrease in tensile strength and flexural stiffness.

Throughout the recycling steps, the strain at break and impact strength increased for carbon and flax fiber filled polypropylene. In contrast, the strain at break and impact strength decreased for the glass fiber polypropylene. This opposite behavior could be attributed to the difference in failure mechanisms. The fiber lengths for carbon and flax fiber reinforced polypropylene are smaller than the critical fiber lengths, resulting in a fiber pull-out failure mechanism. The glass fiber filled polypropylene has a low critical fiber length fiber, causing a fiber fracture failure mechanism.

The addition of flax fibers to polypropylene only results in a small increase of mechanical properties. However, the recyclability of flax fibers is very high as the properties remain almost constant throughout all of the mechanical recycling steps. The recyclability of carbon fiber reinforced polypropylene is lower as the initial stiffness and tensile strength decrease. However, the strain at break and impact strength increase. Glass fiber reinforced polypropylene shows the lowest recyclability as all mechanical properties decrease after ten recycling steps.

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