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Influence of the Recycling Process Parameters on CFRTP Waste Properties

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ABSTRACT

Nowadays, the combination of continuous fibers and thermoplastic polymers as the matrix to continuous fiber-reinforced thermoplastics (CFRTP) is receiving increasing attention due to their potential advantages such as excellent weight-specific mechanical properties, short cycle times, storability, repeated meltability, good formability and the use of alternative joining processes enabling automated large volume manufacturing processes which allow various applications in different industries including transportation, construction among others. As the production rate of these materials increases, the amount of waste for disposal increases, for which recycling strategies need to be established to ensure the sustainability of CFRTP. Hence, these recycling strategies must be developed and evaluated economically and ecologically to close the loop and achieve a circular economy to process recycled fiber-reinforced pellets from CFRTP waste to valuable products e.g., by injection molding. This study presents a mechanical recycling approach from CFRTP waste to injection molded test specimens and evaluates the impact of the individual recycling steps along the recycling chain on the fiber length as the fiber length is detrimental to the resulting mechanical properties. First, the CFRTP waste processability is investigated and conditions for size reduction by cutting and shredding into feedstock for extrusion are defined. Second, fiber-reinforced pellets are produced by twin-screw extrusion. The fiber volume content and the process parameters screw speed and temperature during compounding are varied and the influence of these parameters on the fiber length is determined. Third, the extruded pellets are further processed by injection molding. Here, the influence of screw speed, back pressure, and processing temperature as well as the initial fiber length in the extruded granules on the resulting properties is investigated. Quantitative correlations between material properties and processing parameters are presented and suggestions for gentle processing during recycling are given.

Index Terms *–*Continuous fiber-reinforced thermoplastics, Recycling, Fiber length

1. INTRODUCTION

Continuous fiber-reinforced thermoplastics (CFRTPs) - consist of continuous reinforcing fibers and a thermoplastic matrix polymer. The market for long and continuous fiber-reinforced thermoplastics is growing as these materials have excellent mechanical properties and other advantages such as short cycle times, storability, repeatable meltability, and formability. Unlike thermoset composites, this class of materials is recyclable [1-4]. All these properties enable diverse applications in various industries such as transportation, construction, aerospace, electricity, electronics, biomedical, etc [5,6]. Currently, the value chain for composites is linear and the main disposal routes for composites are co-processing in cement factories or landfilling. The disposal routes for these kinds of materials can be classified into landfilling and incineration, which are not environmentally friendly, as well as recycling. Recycling can be further divided into thermal, chemical, and mechanical recycling [7,8]. Recovering and reusing the reinforcing fibers in composites is the main aim of thermal recycling. The polymer matrix is broken down into molecules of lower molecular weight by heating the composite parts above the decomposition temperature of the polymer. The thermal recycling method can be applied in various ways including pyrolysis, fluidized bed, or microwave-assisted pyrolysis [7]. In chemical recycling, the polymer matrix is removed by solvolysis or dissolution in a suitable solvent. In solvolysis, reactive solvents break down the covalent bonds of the polymer matrix, whereas, in dissolution, the thermoplastic matrix is physically dissolved in a solvent. Chemical recycling can be carried out at lower temperatures in comparison to thermal recycling, as a result, less damage to the recovered fibers occurs. Another advantage of chemical recycling over the thermal recycling process is that it is able to recover both components [7]. A promising method for the recycling of CFRTP is mechanical recycling. Both the reinforcement fibers and the thermoplastic matrix can be recovered and reused through mechanical recycling [9]. In mechanical recycling, the CFRTP part is cut into smaller pieces in several steps to provide a homogeneous quality as well as to achieve processability for the following processing steps. The fiber-reinforced thermoplastic granules are then produced using a twin-screw extruder or an internal mixer, which subsequently can be further processed by injection molding [10]. The main limitation of mechanical recycling is the difficulty to preserve the fiber length and therefore, maintain the properties of the recycled materials. To tackle this problem, optimization of the parameters of mechanical recycling for maximization of the fiber length after re-processing is necessary.

2. MATERIALS AND METHODS

2.1 Materials

2.1.1 Polymer

The polymer, which was used as a matrix material, was the Polypropylene DuPure® SR76 from Ducor. This Polypropylene is typically used for injection molding. Differential scanning calorimetry (DSC 204 F1 Phoenix, NETZSCH-Gerätebau GmbH, Germany) with a heating rate of 20 K/min was used to determine the melting and crystallization temperature of this material, see Table 1. To investigate the flow behavior, a rotational rheometer (Modular Compact Rheometer MCR 302, Anton Paar GmbH, Germany) with a cone-plate setup was used. Figure 1 shows the viscosity for different temperatures. According to the measurements, this polymer illustrates a shear thinning behavior at higher shear rates.

Table 1. Overview of the polymer used in this work.

	Ducor DuPure® SR76
Melt flow rate (230°C / 2.16 kg)	15 g/10 min
Melting temperature	176.4 °C
Crystallization temperature	110.3 °C

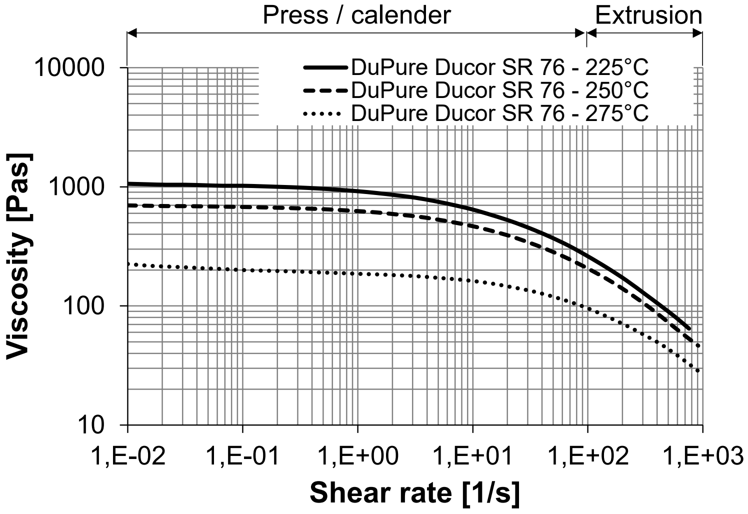


Figure 1. Viscosity data for different temperatures [11].

2.1.2 Reinforcement

A plain weave glass fiber fabric supplied by Porcher Industries Germany GmbH was used as the reinforcing component. The rovings were made of E-glass filaments with a diameter of 9 µm and had a fabric weight of 390 g/m². The fabric was coated with a Volan chromium complex finish (FK144) and had a finishing content between 0.08 and 0.28 %.

2.2 Methods

To evaluate the influence of different parameters on fiber shortening during the mechanical recycling process, there are three main stages for preparing the specimens including mechanical crushing, compounding, and injection molding. It is required to measure the fiber length after each recycling step for various parameters. The respective samples are calcined in a muffle furnace and the remaining fiber content is evaluated regarding the fiber length. The mean values of the evaluation images are calculated by number and weight. The number average fiber length equals the arithmetic mean, while the weight average fiber length put weight on the long fiber portions, as these largely determine the mechanical properties.

2.2.1 Mechanical crushing

For mechanical size reduction, the CFRTP is pre-cut by hand using scissors into approximately 10 cm x 10 cm blanks. Then, the Rapid Granulator AB G200-24K granulator shown in Figure 2 is used for further particle size reduction and mechanical crushing of the CFRTP. After this step, the range of final particles is between 0.5-10 mm.

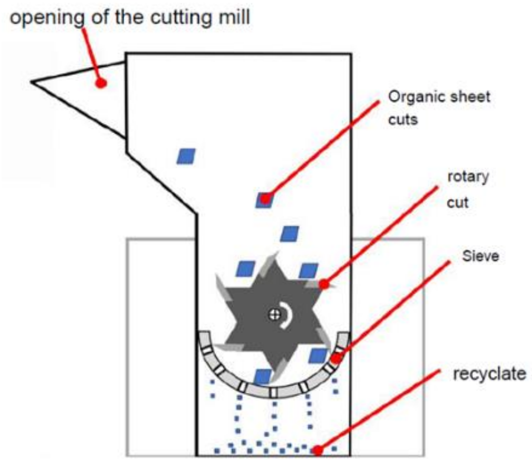


Figure 2. Schematic representation of the granulator.

2.2.2 Compounding

Size-reduced fiber-reinforced thermoplastics are then processed into granules for injection molding using a twin-screw extruder varying three process parameters including fiber volume content, screw speed, and processing temperature. Based on the findings from the literature, the parameters mentioned are expected to be significant influencing variables [12-17]. For this purpose, the crushed CFRTP and new granulates are mixed in a twin-screw extruder, thus producing new fiber composite granulates. Due to the high fiber volume content, the viscosity of the produced fiber-reinforced thermoplastics is high, and thus, dilution with virgin polymer resin is necessary. A total of nine different batches are produced for the preliminary tests. All batches including the processing parameters are listed in Table 2 below.

Table 2. Overview of the extrusion batches of the preliminary tests including processing parameters.

	Fiber volume content	*Processing temperature*	*Screw speed*
Batch E1	5%	210 °C	318 rpm
Batch E2	5%	210 °C	456 rpm
Batch E3	15%	210 °C	318 rpm
Batch E4	15%	210 °C	456 rpm
Batch E5	5%	250 °C	318 rpm
Batch E6	5%	250 °C	456 rpm
Batch E7	15%	250 °C	318 rpm
Batch E8	15%	250 °C	456 rpm
Batch E9	10%	230 °C	387 rpm

2.2.3 Injection molding

The test specimens are produced using an injection molding machine of KraussMaffei with the model designation CX 80-380. A total of eight different batches, for which different process parameters are used, are produced. One process parameter is the initial fiber length. For this purpose, batch E4 from compounding with an initial fiber length of 260 µm (fiber volume content: 19%) and batch E5 with an initial fiber length of 455 µm (fiber volume content: 9%) are used. Furthermore, the back pressure is varied between 20 bar and 60 bar. As in compounding, the screw speed is also changed during the injection molding trials. Settings of 100 rpm and 200 rpm are applied. All batches including the varied processing parameters are listed in Table 3

Table 3 Overview of the injection molding batches of the main investigations including processing parameters.

	Initial fiber length	*Back pressure*	*Screw speed*
Batch S1	260 µm	20 bar	100 rpm
Batch S2	260 µm	20 bar	200 rpm
Batch S3	260 µm	60 bar	100 rpm
Batch S4	260 µm	60 bar	200 rpm
Batch S5	455 µm	20 bar	100 rpm
Batch S6	455 µm	20 bar	200 rpm
Batch S7	455 µm	60 bar	100 rpm
Batch S8	455 µm	60 bar	200 rpm

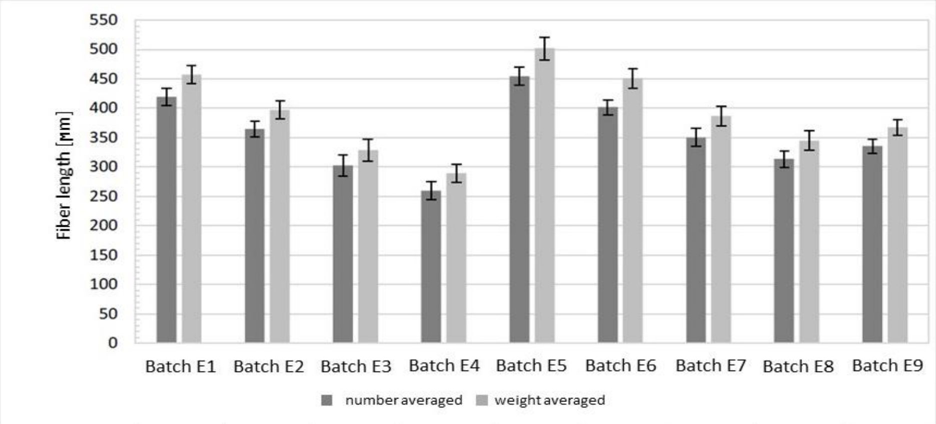
2.2.4 Fiber length evaluation

The resulting fiber lengths are evaluated digitally using an optical microscope from Carl Zeiss AG with the model designation Stemi 2000-C. During the processing of fiber-reinforced plastics, the length of the fibers is reduced in various processing steps due to mechanical influences. The consequences of these influences on the resulting fiber length shall be clarified by evaluating the fiber length distribution. In the first step, the fiber composite sample is calcined in a muffle furnace at 550 °C for 60 minutes to extract the reinforcing fiber. In the next step, part of the fiber sample is placed on a slide and carefully distributed using decanol as dispersing agent. Then the samples are evaluated using optical microscopy and the fiber lengths are statistically determined. The arithmetic mean is calculated for each batch.

3. RESULTS AND DISCUSSION

3.1 Fiber length distribution after compounding

The fiber length distribution is evaluated as described in Section 2.2.4 using an optical microscope. Figure 4 shows the number and weight average fiber lengths including deviations of the individual batches.



T [°C]	210	210	210	210	250	250	250	250	230
φ_f [%]	9	9	19	19	9	9	19	19	14
n_s [min^{-1}]	318	456	318	456	318	456	318	456	387

Figure 3 Number and weight average fiber lengths including deviations of the extruded batches and process parameters.

The measured values show that the weight average fiber length is consistently higher than the number average fiber length for all batches. The lowest deviation is recorded for batch E3 with a percentage deviation of 8%. On the other hand, the largest deviation of the measured values can be seen for batch E6 with a value of 11%. When considering the percentage deviations, these are similar for weight and number average fiber length and all batches. Therefore, only the number average fiber length is considered for further evaluation. The number average fiber lengths, depending on the process parameters, are between 260 µm (batch E4) and 455 µm (batch E5).

3.2 Influence of the fiber volume content on the fiber length after compounding

When considering the influence of the process parameters, it can be said that a higher fiber volume content generally results in a more distinct reduction in fiber length. This is due to the higher fiber-fiber interaction. These results coincide with the findings of *Brast* and *Albrecht*. In both studies, it was observed that in areas with a high fiber volume content strong fiber-fiber interactions were present. [12, 13] In Figure 4, all batches are listed for comparison and the percentage deviation based on the higher measured value observed for a fiber volume content of 9% is calculated. The percentage deviation for these measured values is between -29% and -22%. Thus, a strong influence of the fiber volume content on the resulting fiber length can be identified.

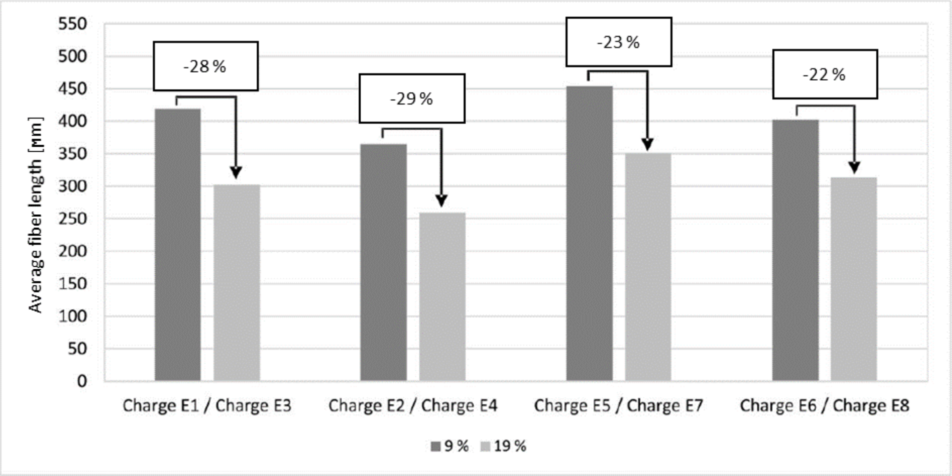


Figure 4 Influence of the fiber volume content in the extrusion process on the resulting fiber length.

3.3 Influence of the process temperature on the fiber length after compounding

The measurement results show a significant shortening of the fiber length when the temperature is decreased. A lower barrel and material temperature leads to a larger melting zone. Since damage to the fibers occurs predominantly in the melting zone, this causes a shortening of the fiber length. These results correspond to the findings of *Brast*. [12] In Figure 5, all batches are listed in relation to the barrel temperature, and the percentage deviation is calculated in relation to the higher measured value. The percentage deviation in the measured values is between 8% and 17%. An influence of the processing temperature can thus be identified.

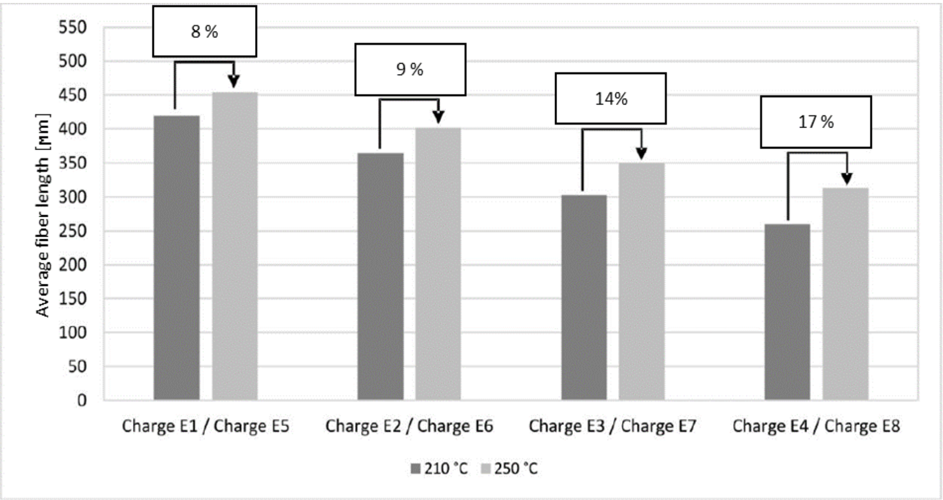


Figure 5 Influence of the processing temperature in the extrusion process on the resulting fiber length.

3.4 Influence of the screw speed on the fiber length after compounding

In Figure 6, all batches are listed in relation to the screw speed change of 138 min-1, and the percentage deviation is calculated in relation to the higher measured value. The percentage deviation of the measured values is between -11% and -14%. Thus, a medium influence of the screw speed can be determined. As depicted in Figure 6, an increase in the screw speed leads to a reduction in the average fiber length. One of the reasons for this is that a higher speed results in greater fiber-extruder interaction. In addition, the fibers in the polymer are exposed to greater movement, resulting in greater fiber-fiber interaction. In conjunction with the higher screw speed, the shear rate increases, which also increases the fiber-matrix, the fiber-fiber, and the fiber-extruder interactions. These observations are similar to the findings by *Yilmazer*. *Yilmazer* discovered that increasing the screw speed of a twin-screw extruder from 250 rpm to 350 rpm leads to a reduction in fiber length. Specifically, when the screw speed was increased by 100 rpm, a fiber length reduction of 7% at a throughput of 70 kg/h and a fiber length reduction of 9% at a throughput of 80 kg/h were observed. [17]

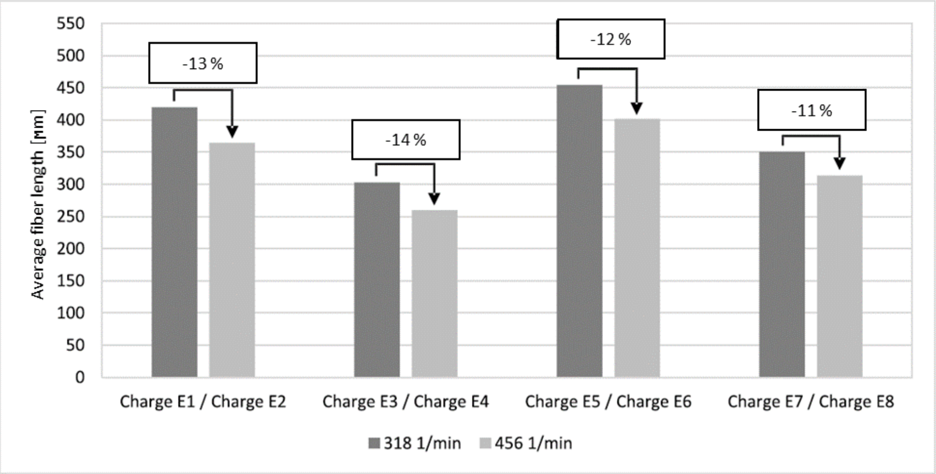
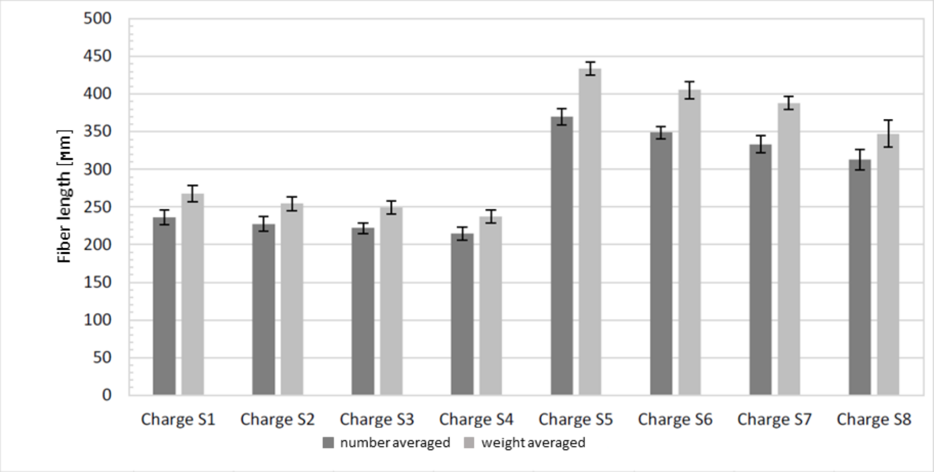


Figure 6 Influence of the screw speed in the extrusion process on the resulting fiber length.

3.5 Fiber length distribution after injection molding

In addition to examining the compounding process with a twin-screw extruder, the influence of different process parameters in the injection molding process is also being considered. Here, the granules produced by twin screw extrusion were used to produce fiber-reinforced test specimens with an injection molding machine. Batch E4 and batch E5 from the twin screw extrusion process served as the basis. Thus, in the production of the injection molded specimens, the influence of the fiber volume content, the initial fiber length, the back pressure, and the screw speed on the resulting fiber length were investigated. A total of eight different batches was produced. Similar to the extrusion process, the fiber length distribution is evaluated by optical microscopy. Figure 7 shows the number and weight average fiber lengths including deviations of the eight injection molded test specimens. The measured values show that the weight average fiber length is consistently higher than the number average fiber length for all batches. The lowest percentage deviation can be seen for batch S8 with a deviation of 10%. The largest percentage deviation is recorded for batch S5 with a value of 15%.



$l_a [\mu m]$ / φ_f [%]	260 / 19	260 / 19	260 / 19	260 / 19	455 / 9	455 / 9	455 / 9	455 / 9
$p\ [bar]$	20	20	60	60	20	20	60	60
$n_s\ [min^{-1}]$	100	200	100	200	100	200	100	200

Figure 7 Number and weight average fiber lengths including deviations in the injection molding batches and process parameters.

3.6 Influence of the initial fiber length on the final fiber length after injection molding

In Figure 8, all batches are listed for comparison and the percentage deviation based on the higher measured value is calculated. The percentage deviation in the measured values is between 31% and 36%. Due to the high influence of the initial fiber length on the fiber deterioration, only the initial fiber length is considered in the following. When comparing the batches in which the initial fiber length was varied, a slight fiber shortening can be seen with a high fiber volume content of 19% and a shorter initial fiber length of 260 µm. However, there is significant fiber deterioration at low fiber volume content of 9% and a longer initial fiber length of 455 µm. This finding is contrary to the influence of the fiber volume content in the extrusion process. However, in addition to the fiber volume content, the initial fiber length differs between the individual batches. The greater fiber deterioration may thus be related to the greater initial fiber length since longer fibers break under lower mechanical loads due to the greater buckling length [18].

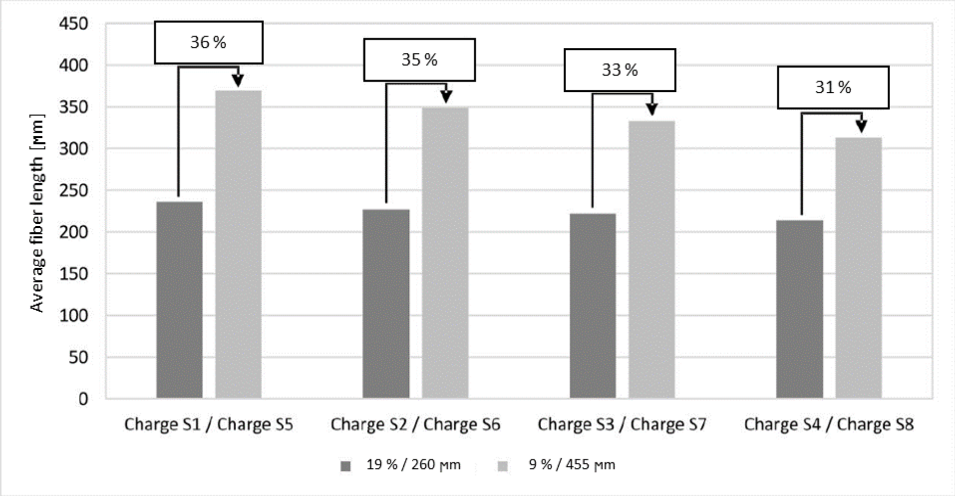


Figure 8 Influence of the fiber volume content / the initial fiber length in the injection molding process on theresulting fiber length.

3.7 Influence of the back pressure on the final fiber length after injection molding

In Figure 9, all batches are listed for comparison and the percentage deviation is calculated in relation to the higher value. The percentage deviation of the measured values is between -6% and -10%. A moderate influence of the dynamic pressure can thus be identified. When considering the influence of back pressure, it can be said that a higher back pressure leads to a reduction in fiber length. Due to the high dynamic pressure, the fiber-plastic melt is thoroughly mixed, resulting in high fiber-fiber, fiber-matrix, and high fiber-machine interactions.

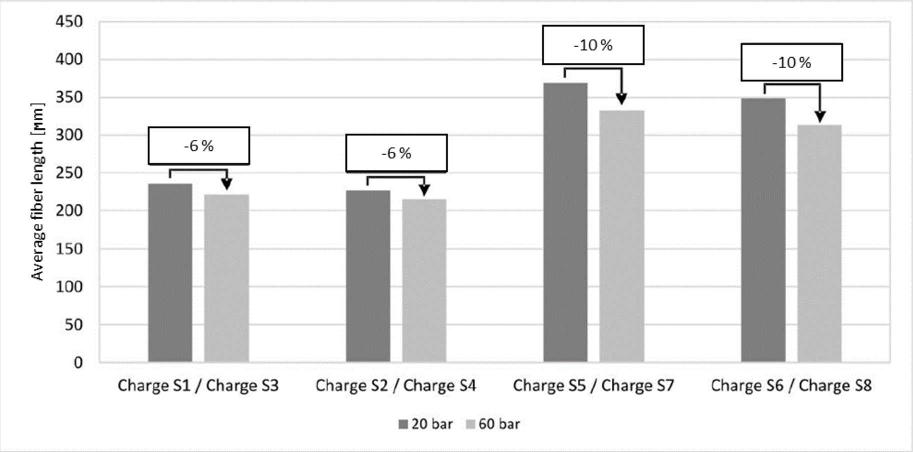


Figure 9 Influence of the back pressure in the injection molding process on the resulting fiber length.

3.8 Influence of the screw speed on the final fiber length after injection molding

All batches in which the screw speed was varied are plotted in Figure 10 for comparison and the percentage deviation based on the higher measured value is calculated. The percentage deviation is between a value of -3% and -6%. Hence, a slight influence of the screw speed on the resulting fiber length can be determined. Increasing screw speed decreases the resulting fiber length. This is due to the higher shear rate and as a result, there is more fiber-machine interaction. Furthermore, the fibers are exposed to faster movement, resulting in greater fiber-fiber interaction. In addition, the higher shear rate causes greater fiber-matrix interaction.

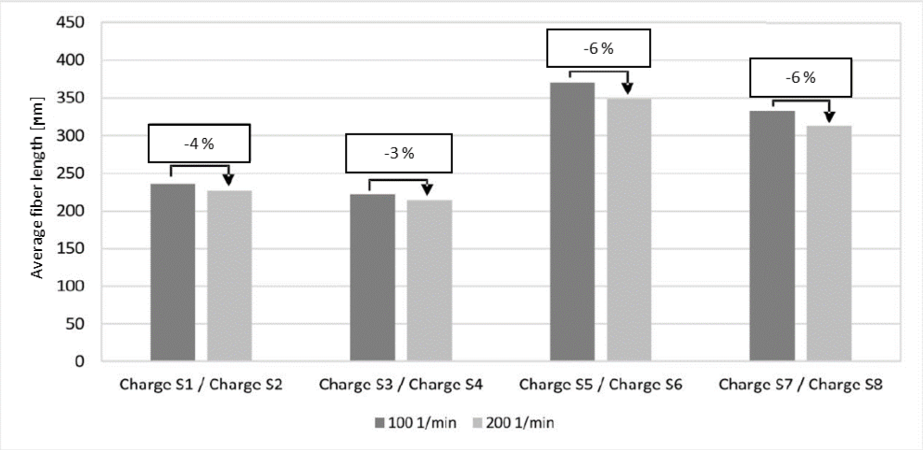


Figure 10 Influence of the screw speed in the injection molding process on the resulting fiber length.

4. CONCLUSION AND OUTLOOK

In conclusion, according to the findings it can be observed that the different recycling steps affect the resulting fiber length after each processing step. During the twin-screw extrusion, the mean fiber lengths of the individual fibers were found to be between 260 µm and 455 µm. The greater fiber volume content has a reducing influence on the resulting fiber length with the average deviation percentage of -25% and thus represents the greatest influence. The higher processing temperature has an increasing influence on the final fiber length with the deviation percentage of 12%, while the higher screw speed has a moderate deviation of -12%. The evaluation of the test specimens after the injection molding process regarding the resulting fiber length showed that the average fiber length depends strongly on the initial fiber length. This means that by increasing the initial fiber length, the final fiber length is increased by a mean deviation of 34%. Because of the longer initial fiber length, there is greater damage to the fibers, but this results in a longer fiber length than with a short initial fiber length. Increasing both back pressure and screw speed, results in a fiber shortening with the average deviation percentage of -8% and -5% respectively. Recycling of CFRTPs is generally possible, but the processing affects the fiber length. Further studies will cover the influence of the recycling steps on mechanical properties.

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