Experimental Study on Fiber Attrition of Long Glass Fiber-Reinforced Thermoplastics under Controlled Conditions in a Couette Flow

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Abstract

Fiber attrition of long glass fiber-reinforced polypropylene in a Couette Flow was studied to obtain a fundamental understanding of the physics in fiber breakage. The developed experimental setup of the Couette rheometer in combination with the developed fiber length measurement technique is able to provide repeatable, accurate and robust data sets to quantify the fiber length reduction during the processing of fiber-filled materials.

This article summarizes the first results of an ongoing experimental study on fiber breakage at the Polymer Engineering Center at the University of Wisconsin-Madison. The impact of fiber concentration, initial fiber length, residence time, melt temperature and processing speed was quantified and studied. The proposed procedure aims to isolate the effect of process variables on fiber breakage. Even for the gentlest processing conditions, the results indicate that the residual fiber length after processing is less than 50% of the initial fiber length. For the most severe processing conditions, the results suggest a reduction to less than 10% of the initial fiber length, which highlights the challenges that fiber breakage poses for processing of long glass fiber-reinforced thermoplastics.

Introduction

Due to their outstanding material properties, lower manufacturing costs and better processability, discontinuous fiber-reinforced polymers have gained importance in industry and are now commonly used in transportation related industries [1–3]. Current trends in the industry aim to use composite materials with longer fibers in order to improve the reinforcing effects for lightweight applications. Injection molding and compression molding of long fiber-reinforced thermoplastics (LFTs) are widely used processes to produce load-carrying parts with advanced mechanical properties. With favorable specific stiffness and impact strength, LFTs can potentially replace traditional materials and decrease the overall weight of automobiles.

As for all fiber-reinforced composites, the final state of the fibers greatly impacts the local and global properties of the molded part [4,5]. During mold filling of fiber-reinforced materials, the configuration of the fiber is significantly changed, reflected in the form of fiber attrition, excessive fiber orientation, fiber jamming and fiber matrix separation [6,7]. In particular, the mechanism behind fiber attrition during processing is not yet fully understood [2]. Along all stages of LFT processing, the length of the fibers decreases substantially, which has a significant impact on the performance of the finished part [5,8]. In general, it is postulated that fiber breakage can be attributed to the following mechanisms [1]:

- Melting effects: The damage mechanism appears at the beginning of the plasticating stage when the pellets are partially fused and the solid-fiber interaction leads to bending stresses within the fibers.
- Fiber-fiber interaction: In highly filled systems, the fibers collide excessively during processing resulting in length reduction due to the interaction between fibers.
- Fiber-melt interaction: Fibers are embedded in the thermoplastic matrix, which introduces large stresses to the fibers due to the hydrodynamic forces during processing.
- Fiber-wall interaction: Fibers collide and interact with the solid walls of the equipment. Abrasion and friction between the fibers and the walls lead to stress concentrations.

A major challenge has been, and remains to be, the availability of adequate measurement techniques that allow accurate fiber attribute measurements of sufficiently large samples in a timely manner [7,9]. Due to the limitations of efficient and accurate characterization methods, the phenomena of fiber breakage has still not been fully understood.

In 2005, Shon et al. studied the breakage of glass fibers in a twin screw extruder and proposed a simple kinetic model for breakage [10]:

$$\frac{dL}{dt} = -k_f(L - L_\infty)$$

where L is the fiber length at time t and k_f is the fiber breakup rate constant. L_∞ is an asymptotic value describing the length at which no further breakage occurs

at the given flow conditions (unbreakable length). By solving the differential equation, the fiber length reduction over time can be obtained, which describes an exponential decay of the fiber length

$$L(t) = (L_0 - L_\infty)\, e^{-k_f t} + L_\infty$$

where L_0 is the initial length at $t = 0$.

The implications of Shon's proposed model are shown in Figure 1, which illustrates the fiber length degradation for a generic sample from 15 mm to 4 mm. The simplicity of the model would allow one to easily estimate the fiber length at a given point in time. However, the underlying physics of two model parameters k_f and L_∞ have not been investigated yet. Shon et al. did not specify how these parameters might be obtained and solely used them as fitting parameters [10].

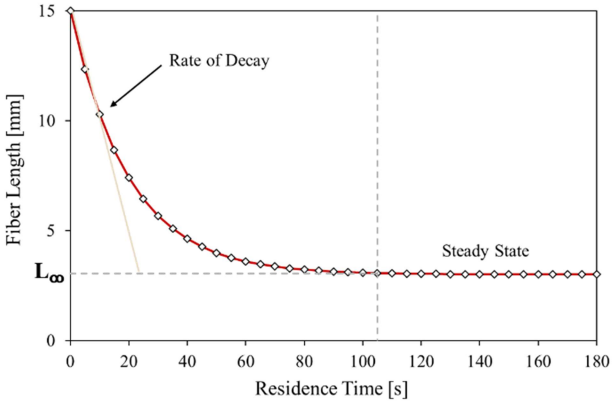

Figure 1. Illustration of expected fiber length reduction over time.

This article presents a proof-of-concept for using a Couette rheometer setup to study fiber breakage under highly controlled conditions. It summarizes the first results of the ongoing experimental study to determine the impact of processing conditions and fiber properties on residual length after processing. The experimental set-up is conceptualized to isolate the effects that drive fiber breakage in processing long glass fiber-reinforced thermoplastics. Ultimately, the experimental study aims to establish a theoretical link between processing conditions and fiber breakage in order to develop adequate predictive tools. While the obtained results reported in this article might not necessary represent fiber lengths in injection molded parts, the study of fiber breakage under controlled conditions is crucial in obtaining a fundamental understanding of the underlying physics in fiber breakage in LFT materials.

Experimental Setup

The fiber breakage mechanism is studied by exposing the fiber-reinforced material to a simple shear under defined conditions in a Couette rheometer. A specifically designed experimental setup for LFT materials was developed at the Polymer Engineering Center (PEC), University of Wisconsin-Madison [11]. The basic concept is illustrated in Figure 2. The Couette rheometer consists of concentric inner and outer cylinders with an annular gap in-between, where the material is placed. While the inner cylinder rotates at a defined speed Ω (or torque T), the outer cylinder is stationary, creating a simple shear flow within annular gap. The temperature is controlled by a set of heater bands and thermocouples. The dimensions of the Couette are characteristic of the conditions during plasticating in injection molding. The length of the annual gap is 80 mm, the inner diameter R_i is 17.5 mm and the outer diameter R_o is 22.5 mm.

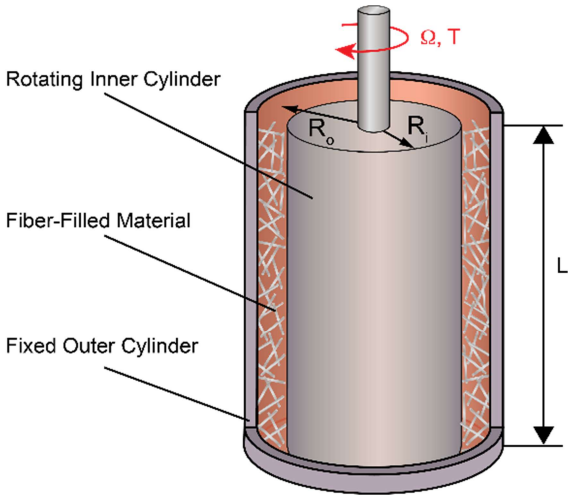

Figure 2. Illustration of the Couette rheometer setup for the study of fiber breakage [12].

Materials and Experimental Plan

The material used in this study is a commercially available glass fiber-reinforced polypropylene (PP) (SABIC®, STAMAX™). The material was supplied as coated fiber pellets with a nominal length of 15 mm. The matrix and fiber type were identical for all experiments of this study and only the glass fiber concentration in the pellets was changed. All experiments were conducted under isothermal conditions to neglect the melting effects of the pellets or impact of changes in viscosity.

The first part of the experiment explored the length degradation over time for varying processing speeds while the fiber concentration and melt temperature (T_M) were kept constant (Table 1). This set of experiments allowed the characterization of length reduction as a function of residence time, t_r, of the material exposed to the Couette flow. Hence, the rate of fiber breakage can be evaluated and the impact of processing conditions on k_f and L_∞ can be quantified.

Table 1. Experimental plan 1: Fiber length reduction over time.

Property	Value
Fiber concentration [%wt.]	30
Melt temperature T_M [˚C]	250
Rotation per minute [rpm]	50, 100 and 150
Residence time t_r [s]	20 to 300

The second part of this experimental plan aimed to study the impact of various variables on the steady state fiber length (unbreakable length, L_∞) for large strains. For each variant of this plan, the residence time and processing speed were selected to ensure that L_∞ was reached based on the results of the first experimental plan. Table 3 summarizes the process variables studied for this approach.

Table 2. Experimental plan 2: Evaluation of various process variables on the unbreakable length L_∞ .

Variable	Variants
Fiber concentration [%wt.]	20, 30 and 40
Melt temperature T_M [˚C]	220 and 280
Initial pellet (fiber) length	3, 6, 9, 12 and 15

The fiber concentration was varied by processing different SABIC® STAMAX™ pellets (20YM240, 30YM240 and 40YM240). The matrix and fibers are identical for these trials and only the concentration of glass fibers in the pellets was varied. It is expected that an increase in fiber concentration causes elevated fiber breakage due to more fiber-fiber interactions in the suspension. The melt temperature has a direct influence on the viscosity of the suspension and, thus, on the stresses that the fibers are exposed to. Lastly, the initial fiber length was varied by manually cutting pellets into lengths between 3 mm and 15 mm. Overall, the results presented in this article summarize an exploratory study of the influencing factors in fiber breakage and do not yet resemble a full factorial experimental study.

Fiber Length Measurement Technique

The characterization of the fiber length distribution of fiber-filled materials is a cumbersome task since even small samples can be comprised of millions of fibers. A wide variety of measurement approaches exist, but no industry standard has been defined yet [7,9]. The substantial differences in the measurement concepts and lack of a standard approach raise questions about repeatability as well as comparability of the results, as pointed out in [7].

At the PEC, a new measurement technique was developed using a time-efficient dispersion system as well as a fully automated image-processing algorithm to measure a large amount of fibers without needing manual fiber dispersion or detection. To ensure that the measured fiber length distribution is statistically representative, at least 20,000 fibers were analyzed for each sample. The measurement procedure is described in detail in [7].

For each experimental variation, three specimens were manufactured in the Couette rheometer. From each specimen, at least three samples were extracted and measured. Consequently, at least nine measurements were performed for each data point of the experimental study. The raw output of the measured samples was processed statistically and is depicted as the cumulative distribution or characteristic average values (number and weight average fiber length). The number average L_n is defined as follows

$$L_n = \frac{\sum N_i l_i}{\sum N_i}$$

where N_i is the number or frequency of fibers with the length l_i. The average weight length, L_w, emphasizes the proportion of long fibers in the population by weighting. It is defined as

$$L_w = \frac{\sum N_i l_i^2}{\sum N_i l_i}$$

An alternative to describe the characteristic length of a fiber population is the cumulative fiber length distribution $\mathfrak{W}$, which is computed as [1]

$$\mathfrak{W}(l) = \frac{1}{\mathfrak{L}} \int_0^L \mathfrak{w}(l) dl$$

where $\mathfrak{w}(l)$ is the fiber length distribution, $\mathfrak{L}$ is the total fiber length if the sample and l is the length of individual fibers.

Results

The results are presented in four subsections, each isolating one of the process variables and its effect on fiber breakage. The first section focuses on the length reduction as a function of residence time for different processing speeds. Subsequent sections each summarize the obtained results on the impact of temperature, fiber concentration and initial fiber length on the residual fiber length for long residence times.

Fiber Length Reduction over Time

The residual fiber length at residence times between 20 s and 300 s was measured at three different processing speeds (50, 100 and 150 rpm). The markers indicate the corresponding average length including the standard deviation represented by error bars. Additionally, Shon's model was fitted to the data sets (solid lines) and the model parameters are summarized in Table 3.

Figure 3 shows the reduction of L_n and L_w as a function of the residence time for a melt temperature of 250 ºC, a fiber concentration of 30%wt, and a processing speed of 50 rpm. For these processing conditions, the fiber

weight average L_w was reduced from 15 mm down to 1.6 mm after a residence time of 300 s. Overall, the results confirm the expected exponential decay with a strong length reduction rate between 0 s to 100 s of processing. The unbreakable length was reached at a residence time between 120 s and 180 s. Figure 4 and Figure 5 summarize the measurements for a processing speed of 100 rpm and 150 rpm respectively. The results suggest the same trend in length reduction. As expected, the increased processing speed accelerates the rate at which the fibers break and also decrease L_∞.

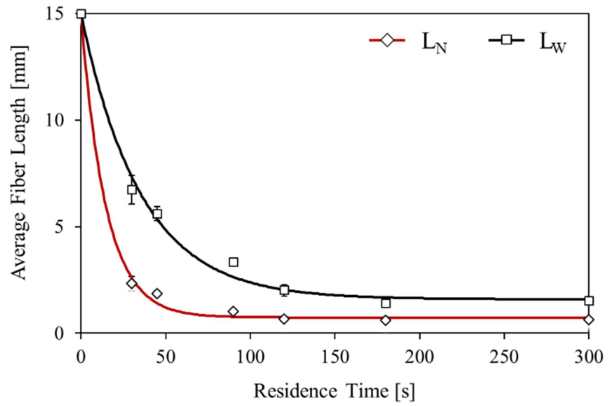

Figure 3. Fiber length reduction over time measured in the Couette Rheometer for: 30%wt., 50 rpm and 250 ºC. The solid lines show the fitted Shon Model.

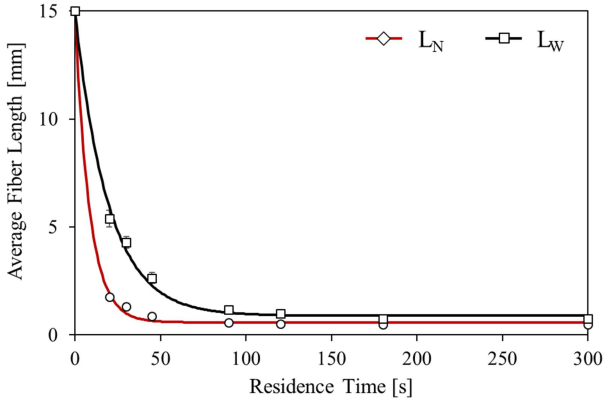

Figure 4. Fiber length reduction over time measured in the Couette Rheometer for: 30%wt., 100 rpm and 250 ºC. The solid lines show the fitted Shon Model.

Table 3. Fitted Shon model parameters for L_n and L_w

Processing Speed	$L_{n,\infty}$ [mm]	$k_{n,f}$ [-]	$L_{w,\infty}$ [mm]	$k_{w,f}$ [-]
50 rpm	0.74	0.07	1.60	0.03
100 rpm	0.58	0.12	0.89	0.05
150 rpm	0.51	0.15	0.78	0.07

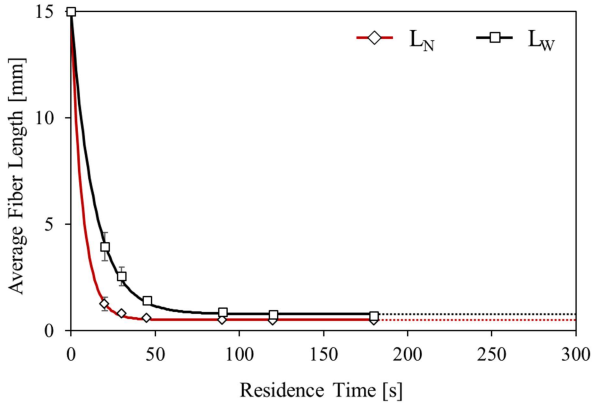

Figure 5. Fiber length reduction over time measured in the Couette Rheometer for: 30%wt., 150 rpm and 250 ºC. The solid lines show the fitted Shon Model. (The dotted lines indicate the expected unbreakable length L_∞ for longer residence times).

Figure 6 shows the comparison of the fitted curves for L_w. The graph clearly shows the accelerated fiber breakage rate at higher processing speeds. The unbreakable length for 150 rpm is 0.78 mm, which is about 50% of L_w at 50 rpm. However, the difference between 100 rpm and 150 rpm is only 14% suggesting a decreased impact of the processing speed on the unbreakable fiber length.

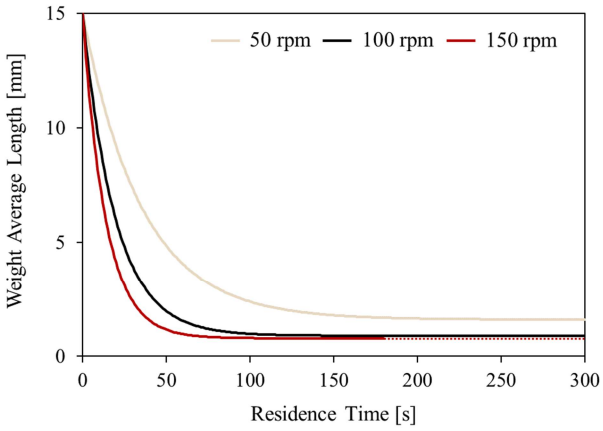

Figure 6. Comparison of reduction of L_w over time for 50, 100 and 150 rpm (250 ºC and 30%wt.) illustrated with fitted Shon's model.

Impact of Melt Temperature

Due to the temperature dependency of the suspension viscosity, the temperature influences the hydrodynamic stresses that the fibers are exposed to. Hence, Couette rheometer experiments are performed to quantify the impact of melt temperature on the residual fiber length. Figure 7 and Figure 8 show the residual fiber length for 220 ºC and 280 ºC for residence times of 120 s and 300 s respectively. These temperatures are the upper and lower bounds for processing this material according to the material supplier [13]. Hence, the obtained measurements represent the most severe impact of temperature that can be expected for the given fiber concentration and processing speed. At a residence time of 120 s, the measurements

suggest that the increased suspension viscosity leads to reduction of L_w from 5.41 mm at 280 ºC to 4.22 mm at 220 ºC, which is a 22.1% difference in the residual length. For 300 s, the measured L_w is 3.03 mm at 280 ºC and 2.13 mm at 220 ºC (29.7% difference).

The average shear rate in the Couette Rheometer at 46 rpm is 19.6 s^{-1}. To estimate the viscosity at 220 ºC and 280 ºC was obtained from the Cross-WLF model fitted to measured data [14]. The suspension viscosity for a shear rate of 19.5 s^{-1} at 220 ºC and 280 ºC is 520 Pa·s and 175 Pa·s, respectively. This shows that the shear stresses in the system are a factor of three higher for the lower temperature, which explains the reduced residual length in experiments.

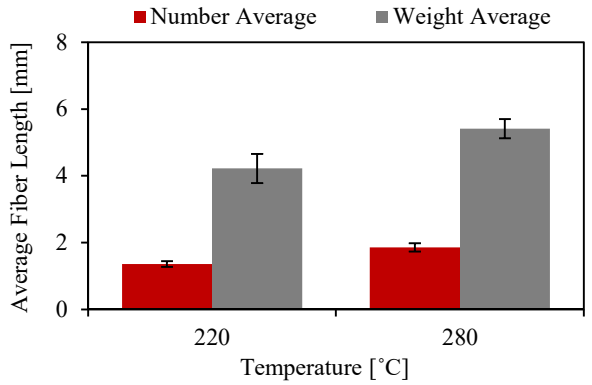

Figure 7. Couette rheometer results: Measured fiber length (L_n and L_w) for 220 ºC and 280 ºC at 120 s residence time (30%wt., 46 rpm).

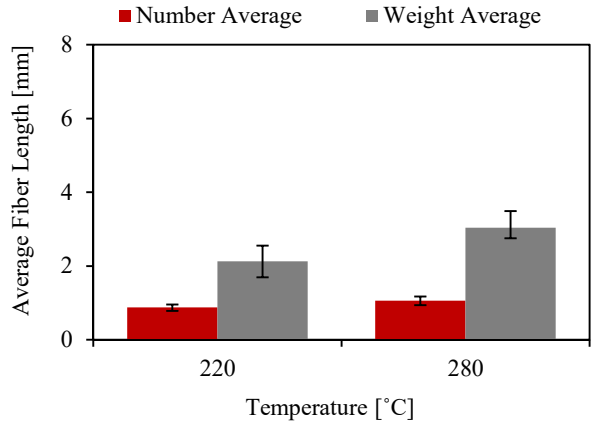

Figure 8. Couette rheometer results: Measured fiber length (L_n and L_w) for 220 ºC and 280 ºC at 300 s residence time (30%wt., 46 rpm).

Impact of Fiber Concentration

At increased fiber concentration, it can be assumed that more fiber-fiber interactions occur. Additionally, the suspension viscosity increases with the fiber concentration [14]. Since both mechanisms can accelerate fiber breakage, it is safe to assume that fiber breakage strongly depends on fiber concentration.

As shown in Figure 9 and , the anticipated dependency between residual fiber length and fiber concentration was reproduced in Couette rheometer experiments for three different fiber concentrations (20%wt., 30%wt. and 40%wt.) at two processing speeds (46 rpm and 160 rpm). At 46 rpm, the results suggest that increasing the fiber concentration by 10%wt. reduces the residual length by 25%. This reduction is elevated for the 160 rpm trials, where an increase in concentration by 10%wt. reduces L_w by 30%.

For a graphical comparison of the measured fiber length distributions, Figure 11 shows the cumulative length distribution for the 46 rpm. The shift of the graphs to the left for increasing fiber concentration, clearly show the elevated fiber breakage. Moreover, the results for the 20%wt. graph suggest that approximately 25% of the fibers are still longer than 7 mm for the given processing conditions. At 40%wt. fiber concentration, only 5% of the fibers are longer than 7 mm.

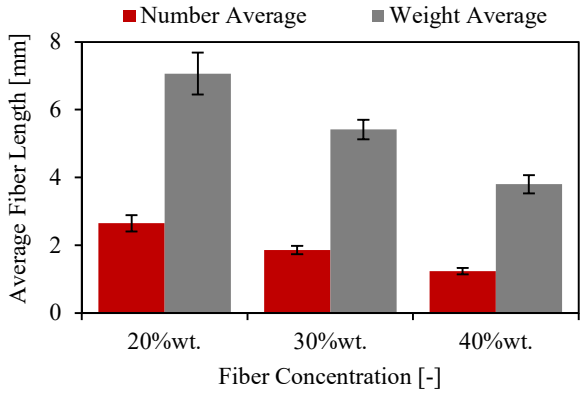

Figure 9. Couette rheometer results: Measured fiber length (L_n and L_w) for 20%wt., 30%wt. and 40%wt. fiber concentration for a processing speed of 46 rpm (120 s and 280 ºC).

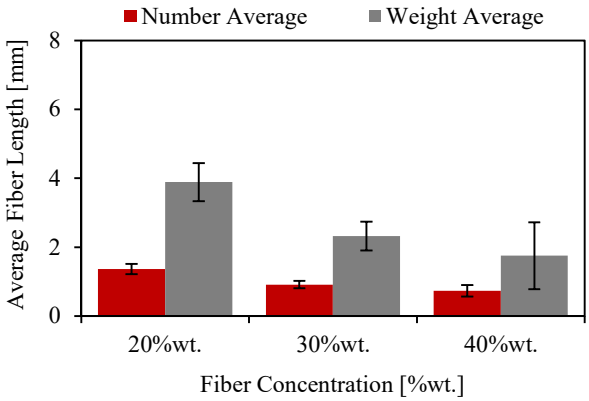

Figure 10. Couette rheometer results: Measured fiber length (Ln and Lw) for 20%wt., 30%wt. and 40%wt. fiber concentration for a processing speed of 160 rpm (120 s and 280 ºC).

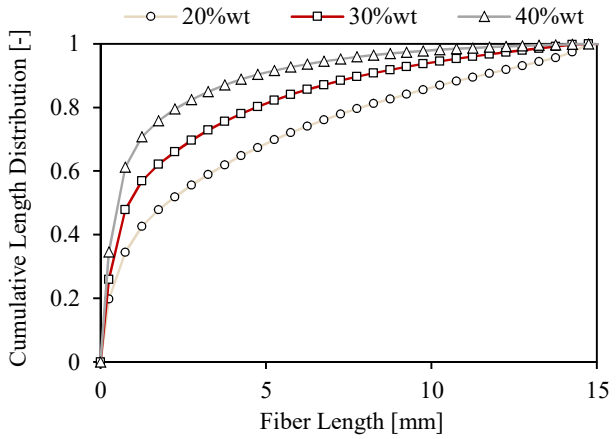

Figure 11. Couette rheometer results: Cumulative length distribution for 20%wt., 30%wt. and 40%wt. fiber concentration (46 rpm, 280 ºC and 120 seconds).

Impact of Initial Fiber Length

In order to evaluate any influence of the initial fiber length on the unbreakable fiber length L_∞, pellets with varying length but identical materials were manufactured manually. The initial length of the fibers was changed in increments of 3 mm up to the original length of 15 mm. The processing conditions were selected to ensure the unbreakable length L_∞ was reached by running the experiments at large strains. Hence, the hypothesis $L_\infty \neq f(L_0)$ can be tested and verified at one processing condition.

Figure 12 and Figure 13 show the measured residual fiber length in terms of L_n and L_w, respectively, for the processing conditions of 100 rpm, 180 s and 250 ºC. The results confirm that the fiber converges to the same length regardless of the initial length of the fibers. These results do not provide any information on fiber breakage during the transient stage (before L_∞ is reached). However, they suggest limitations in solely increasing the length of the LFT pellets to increase the residual length in molded parts.

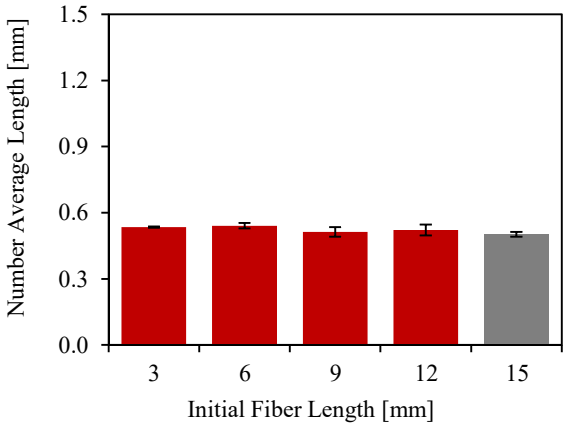

Figure 12. Couette rheometer results: Measured L_n for varying initial fiber length (30%wt., 250 ºC, 100 rpm, 180 s).

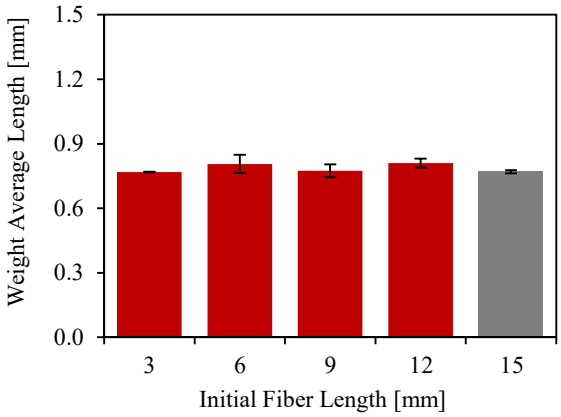

Figure 13. Couette rheometer results: Measured L_w for varying initial fiber length (30%wt., 250 ºC, 100 rpm, 180 s).

Discussion

The presented results of this ongoing experimental study underline the challenge that fiber breakage phenomena pose for long glass fiber-reinforced thermoplastics processing. In the Couette rheometer experiments, it was observed that even for the gentlest processing conditions (20%wt., 46 rpm, 280 ºC and 120 s), the residual fiber length is less than 50% of the initial fiber length. As soon as the fiber concentration or the processing speed is increased or the melt temperature is lowered, the residual fiber length decreases even further. At the most severe processing conditions (30%wt., 150 rpm, 250 ºC and 180 s) the residual length was measured to be 0.78 mm, which is less than 10% of the initial fiber length.

The preliminary results from the Couette rheometer experiments suggest that there might be no impact of the initial length on the unbreakable length. This result is valuable for modeling of fiber breakage, since it implies that the residual fiber length at long residence times converges to an asymptotic value (the unbreakable fiber length L_∞) determined by the material properties and the process conditions.

However, it should be realized that L_∞ obtained from the Couette rheometer experiments does not necessarily represent the residual fiber length observed in injection molded parts, where occasionally larger average lengths were reported [15]. The reason might be that L_∞ has not been reached during injection molding as the residence time at a typical injection molding process is less than 180 s. Consequently, it still has to be investigated how the initial fiber length affects the fiber length reduction over time in the Coeutte rheometer setup. Ultimately, this will support studying the importance of the initial fiber length for the residual fiber length in injection molded parts. This is a particularly important research topic due to the increased use of longer pellets (longer fiber) in LFT materials to achieve longer residual fiber lengths and improve mechanical properties in the molded part [16].

This practice might be limited and longer fibers might not necessarily be achievable by solely increasing the initial length of the fibers.

Overall, the presented results of this study are aligned with the expected fiber breakage mechanism and an exponential decay as a function of residence time was observed. Shon's two parameter model was successfully applied to fit the experimental results of this work. However, the underlying physics and process dependencies of the model parameters still need to be addressed.

Conclusions and Outlook

The experimental setup of the Couette rheometer in combination with the developed measurement technique was proven to be a valuable tool to study fiber breakage of long glass fiber-reinforced thermoplastics under controlled conditions. Using the proposed methodology, a repeatable, accurate and robust set of experimental data can be obtained. The findings are of tremendous value in testing available fiber breakage models, improving their capabilities or developing new modeling approaches. Most importantly, the experimental setup allows one to isolate, study and quantify the impact of process variables.

The ongoing experimental study will provide the experimental data needed to obtain a better understanding of the underlying physics of fiber breakage for LFT materials. Ultimately, the results will be translated to actual LFT processes and improved processing guidelines will be developed to reduce fiber length degradation.

Acknowledgements

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