MECHANICAL CHARACTERIZATION OF JUVENILE AND RECYCLED CARBON FIBERS USING SINGLE FIBER TENSILE TESTS

Jonas Hüther[1], Ina Kohlschreiber[1], Matthias Stoll[1], Kay André Weidenmann[1]

[1]Karlsruhe Institute of Technology (KIT), Institute for Applied Materials (IAM-WK),
Department: Hybrid and Lightweight Materials, Kaiserstr. 12, 76131 Karlsruhe, Germany
Email: Jonas.Huether@kit.edu, https://www.iam.kit.edu/wk/english/index.php

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Abstract

As composite materials are designed in a way that they contain durable connections between at least two different materials, recycling of these structures is challenging. Thereby, recycling may compromise the material's mechanical properties. Nonetheless, to meet the needs of sustainability, recycling is increasingly applied in different fields of industry and research, including composites. Single fiber tensile testing is a valuable method to fully characterize the mechanical properties and to understand the effects of recycling. However, single fiber tensile testing procedures have to be optimized to obtain reliable and comparable data. This work compares two different single fiber tensile testing methods as well as the tensile strength of juvenile and recycled fibers using different recycling routes. Scanning electron microscopy images support the mechanical test and reveal surface changes. Further, A perspective for future optimization in single fiber testing is given.

1. Introduction

One of the driving forces to use fiber reinforced polymers are their outstanding material properties that can hardly be achieved using conventional bulk materials [1].
Here, a contradiction arises: To achieve good mechanical properties, a strong and permanent connection between the reinforcing fibers and matrix material has to be ensured. However, the better the connection, the more difficult it is to separate reinforcement and matrix when reaching the end of life. For most polymeric composites, the fibers are by far more valuable than the polymer matrix as figure 1 reveals.

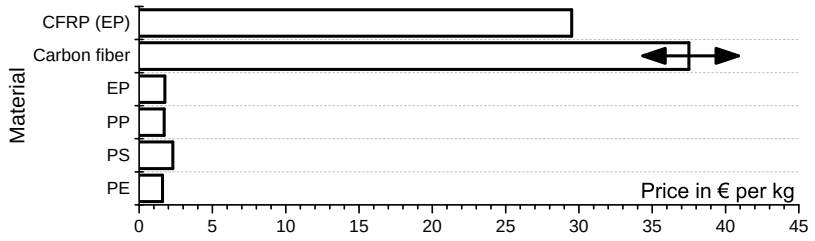

Figure 1. Price per kg for several bulk polymers compared to CFRP and carbon fiber [2]

Thus, it is important to separate the fibers from the polymer and, if possibly, feed them back into inde-

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pendent recycling processes. On commercial scale, only pyrolysis is used, causing the matrix to be burnt of and lost for further uses [3]. However, the fibers can be reused in a new composite.
When discussing recycling in terms of composite materials, for instance in automobile industry, two main categories of recycling processes have to be considered as shown in figure 2.

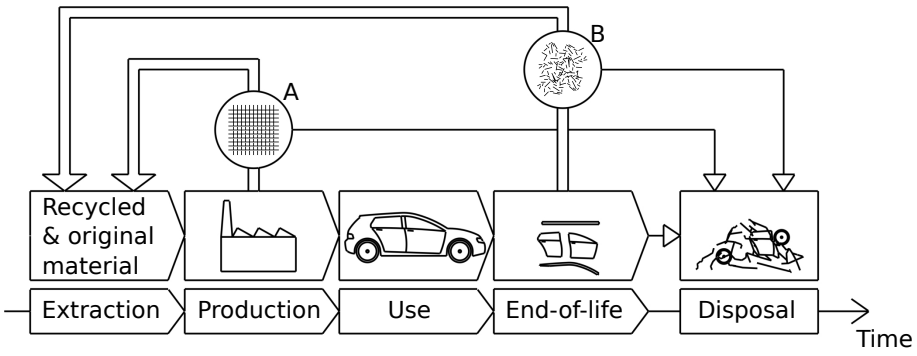

Figure 2. Example case: Material flow in car production. Dry fibers (A) and fibers regained from composites (B), can be used to establish a circular economy.

One is the reuse of dry fiber material left over during a primary production process (figure 2-A). Here, the aim of recycling is to provide a uniform length distribution and constant mechanical properties to feed the material back into a formative production process including impregnation. Albeit the material has not been impregnated with resin, its properties might be impaired due to handling and cutting. The second category comprises fibers already used in composite materials (figure 2-B). To regain the fibers, they have to be separated from the polymeric matrix. Different methods, for instance pyrolysis, microwave pyrolysis, solvents and supercritical fluids, are being researched but all recycling processes stress the materials with a variety of loads (heat, mechanical loads, friction). Thus, the fiber's properties might be significantly altered. [3–5]

Thanks to material circulation higher rates of recycling in production and of end-of-life products are feasible, and less material has to be disposed. Less disposal limits the hazards to the environment and decreases the need for new materials produced via primary processes (see figure2). [1, 6]

It is assumed that recycling affects the material's properties. Compared to newly produced (juvenile) polymers and fibers, they undergo a variety of different influences such as heating and cooling, loads, cutting, bending etc. To test these influences, two different procedures are possible: On the one hand, fibers can be embedded in a new matrix comparing its properties to composites made from new fiber material with the same matrix. On the other hand, single fibers or fiber-bundles can be tested and compared to their juvenile equivalent. [7]

In this paper, single fiber tensile tests are performed. The difficulty of single fiber handling and testing is best visualized by demonstrating the small dimensions (diameters of approximately 5 to 10 μm) as illustrated by figure 3.

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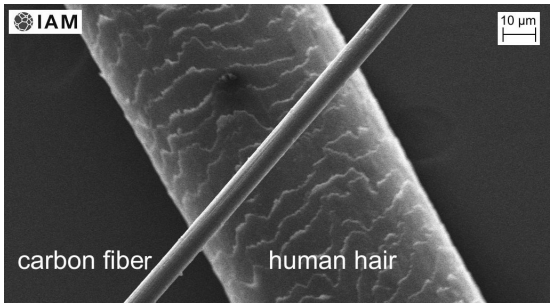

Figure 3. Comparing the diameters of a human hair to a new carbon fiber

2. Materials and methods

As described in the introduction, fibers can be recycled from production processes (see figure 2-A) or from cured and used composite materials (figure 2-B). In this work, dry fiber waste as well as fibers regained from a composite are compared with new fibers. All carbon fibers used in the experimental part were taken from a *Zoltek PANEX 35 stitch-bonded uni-directional carbon fabric*. According to Zoltek's data sheet their tensile strength is 3999 MPa and the tensile modulus 241 GPa.

For each group, one exemplary material is selected and thus the following materials are used in this work:

Table 1. Tested fiber types and specimen overview. Base of all tests was a *Zoltek PANEX 35 fabric*

Type	ID	Description
Juvenile	REF	New fibers
Microwave pyrolysis	MP	Impregnated composite, resin removed by microwave pyrolysis
Granulator	S	Dry carbon fiber fabrics cut by a granulator

Both recycling procedures were run by the Fraunhofer Institute for Chemical Technology (ICT). For the S-samples, a granulator was run applying a sieve with hole size of 20 mm. For MP, microwave pyrolysis was chosen as it is a non-commercial technique. To ease the verification of the tensile testing devices in use, recycling parameters with a significant impact on the fiber quality were chosen. For this treatment, 500 W were applied for approximately 1.5 minutes (cf. [7, 8])

3. Experimental

3.1. Single fiber tensile tests

Single fiber tests were performed commercially at the FaserInsitut BREmen (FIBRE) using a standard single fiber testing procedure and at KIT with a new testing device that is currently being set up. Data for tensile test was evaluated seperately for each set of experiments according to Weibull analysis, correlating failure probability with tensile strengths.

Dia-stron Tensile Tester All tests at FIBRE were performed on a *Dia-stron* tensile testing device combining a *LEX810 High resolution Tensile Tester* and a *FDAS765 High Resolution Dimensional Measurement System*. New carbon fibers were tested with a clamping length of 5, 10, 20 and 30 mm using a standard procedure of FIBRE. For each clamping length, 15 fibers were tested. Due to the loss in length during recycling, the fibers taken from the granulator and the microwave pyrolysis were tested solely with a clamping length of 5 mm and 12 to 15 tests were performed. Cross head velocity for all tests was 0.033 mm/min. Data handed over by FIBRE contained areas measured by means of laser diffraction, tensile strengths, elongations and Young's modulus. Nonetheless, the machine's rigidity is not correlated to the elongations measurement.

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KIT's in-house device SiFiT Several single fiber testing devices exist but none accomplishes all the requirements hoped-for by Karlsruhe Institute of Technology (KIT). These requirements are:

- Direct diameter measurement (independent of density)
- Determination roundness
- Flexible clamping lengths and very short fiber lengths
- Different clamping forces
- Applicability for different kind of fibers
- Correct determination of elongation and Young's modulus
- Low initial forces
- Usability and semi-automation
- Access to all test parameters

Tests according to ASTM D3379-75 using a paper frame were performed at KIT. However, this method is, particularly in case of carbon fibers, complicated due to the time consuming preparation procedure and handling. [9]

To overcome the disadvantages of other single fiber testing devices, a new setup, called SiFiT (Compressed Air Testing Machine), is built at the Institute of Applied Materials at KIT. This in-house solution uses compressed air for the basic functions of the new testing device. Firstly, the compressed air is used to open and close parallel grippers and to fasten the carbon fiber properly. Secondly, using the Venturi principle, a suction is evoked and controls the alignment of the lower end of the carbon fiber. Thus, clamping of the specimens is straightforward and fast with this new device while initial loads on the fiber are kept low. The SiFiT further facilitates a variable clamping length and variable cross head velocities to ensure constant strains for different fiber lengths.

The setup of the SiFiT is displayed in figure 4 and functions as described in the following: The cross head (a) is moved to the required position and the upper end of a fiber (d) is clamped with the upper gripper (c1). By the suction of the Venturi nozzle, the lower end of the fiber is pulled into the nozzle and thereby aligned. The lower end is fixated by the lower gripper (c2). The grippers were designed at KIT and are mounted on a guide bar of type *KTG 50* by *Schunk* for parallel gripping. The nozzle is of type *VN-05-H-T2-PQ1-VQ1-RQ1* by *Festo*. A *VHX-600 (gen II)* digital microscope by *Keyence* is located next to the SiFiT, focusing on the fiber to measure its diameter prior to the tensile testing. To do so, the fiber is aligned by moving the cross head to apply an initial load of 5 MPa. After this, the fibers are tested until fracture with a strain rate $0.02\,min^{-1}$. Thus, cross head velocity varies with different clamping lengths.

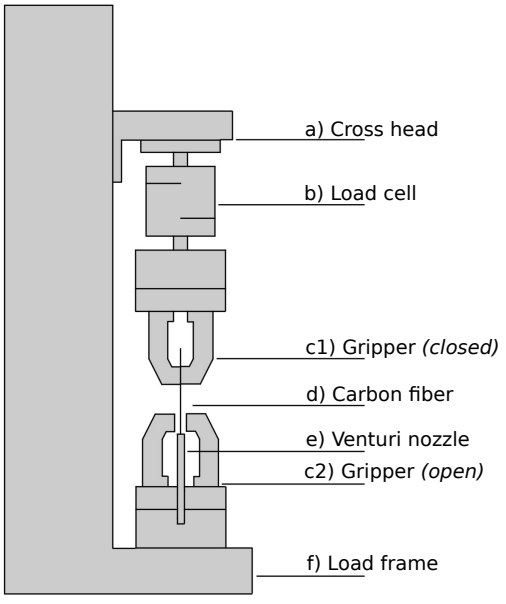

Figure 4. Sketch of the experimental setup for the SiFiT tensile testing device

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3.2. SEM investigations

Scanning electron microscopy was carried out on a Zeiss EVO 500 SEM and it serves two purposes in this work. Firstly, it used to verify the optical diameter measurements used for the mechanical tests. Secondly, the fiber surfaces are investigated to ascertain if the recycling process has a visible influence. For the three types of fibers compared in this paper, SEM-images were taken for a set of magnifications (1000x, 2500x, 5000x, 10000x). The accelerating voltage was set to 5 kV (15 in some cases) and a secondary electron detector was used.

3.2.1. Fiber diameter determination

For diameter measurements, images with magnifications of 2500x and 5000x were chosen. Only fibers in focus were taken into consideration. Magnification was varied due to the following reasons:

a) A higher magnification leads to more accurate results
b) On an image with lower magnification, more fibers are visible. Thus, out-of-roundness and variations in diameter due to production become less significant.

For each type of fiber, at least 35 individual diameter measurements on a variety of fibers were taken.

3.2.2. Surface investigation

The quality of the surfaces were only investigated visually. The images shown in the section results are representatives of the respective materials (REF, MP, S).

4. Results

In the following, results of the SEM observations and tensile strength test are given.

4.1. Diameter measurement

Diameter measurements were taken as described beforehand using SEM images. Further, diameters obtained during the mechanical experiments are listed in table 2. For the FIBRE experiments, measured areas are transferred to diameters mathematically to ease comparison.

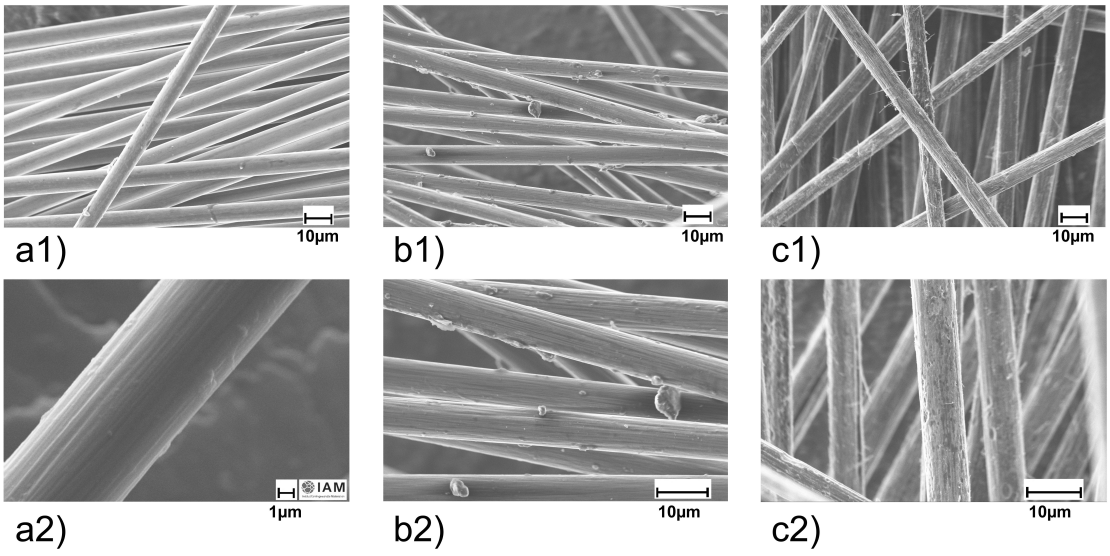

Figure 5. SEM images of juvenile fibers (a), dry fibers recycled in a granulator (b) and with microwave pyrolysis (c).

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Table 2. Diameters measured with different methods

	SEM			Dia-stron			SiFiT
	REF	MP	S	REF	MP	S	REF
diameter in μm	7.41	7.25	7.32	7.10	7.24	7.44	7.39
standard deviation	0.26	0.48	0.44	0.40	1.21	0.48	0.28

4.2. Surface

The juvenile fibers, as represented by figure 5 shows the typical lengthwise surface pattern of carbon fibers [3], [10]. The fibers cut in a granulator show a similar surface, however more particles were found. A variety of particles can be released during the granulating process, such as short fiber filaments, dust and other inpurifications. Recycled carbon fibers regained by microwave pyrolysis appears differently in SEM-images. The typical fiber structure can be observed, however, two phenomenons occur: Firstly, fibrous fragments can be found, leading to the assumption that they might be pealed away from the fiber's surface. Secondly, voids are visible. They could correspond either to burns of the fiber or resin residues. SEM-images show that both investigated recycling processes alter the fiber's surface, but that the dry and cut fibers are by far less affected. However, it is beyond the scope of this work to draw direct conclusions between the fiber surface and its material properties.

4.3. Fiber properties

Results of all mechanical single fiber tensile tests are displayed in table 3 and will be discussed in section 5.

Table 3. Weibull statistics of single fiber tensile test for juvenile and recycled fibers by means of a granulator and microwave pyrolysis

Device	Type	Clamping length	Weibull strength	Weibull modulus
			in MPa	
FIBRE	REF	5	4630,2	7,98
FIBRE	REF	10	4595,7	5,43
FIBRE	REF	20	4619,5	6,60
FIBRE	REF	30	3896,2	5,57
SiFiT	REF	5	3620,2	4,54
SiFiT	REF	10	4433,4	5,71
SiFiT	REF	20	4479,2	5,33
SiFiT	REF	30	4404,9	7,23
FIBRE	MP	5	1979,9	1,75
FIBRE	S	5	5596,7	4,05

5. Discussion

For all sets of tests (different types of fibers, different clamping lengths, different testing devices) 12 to 15 single fibers were tested. Thus, a trend can be noted but the results have to be verified by additional tests. For the juvenile fibers, the total specimen count was higher, but they were grouped according to the clamping lengths.

The tensile strengths of the juvenile carbon fibers tested by FIBRE and by SiFiT were consistent and corresponded to the values given by the manufacturer. SEM investigations showed, as expected, clean surfaces.

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The carbon fibers cut in a granulator show higher tensile strengths than the juvenile ones. This result is not expected and further investigations have to be carried out. The raw data reveals diameters in an acceptable range but forces at fracture at significantly higher values. The cutting process rather delivers a distribution of fiber lengths than a uniform fiber length. As a certain amount of fibers is shorter than the minimum clamping length, therefore some fibers cannot be used for tensile testing. Thus, this recycling process is preselecting the entirety of specimen. As a consequence, a statistically accurate prediction unfeasible. SEM images show particles attached to the cut fibers, but no fiber damage is visible. As the count of specimen was low, a statistically reliable conclusion cannot be drawn.

Tensile test of fibers regained by microwave pyrolysis and tested at FIBRE show a significant decrease in tensile strength. The fiber surface is impurified by different particles and voids. As aforementioned, pyrolysis was, on purpose, performed on a high energy level to demonstrate recycling damage. The fibers are not cut during the recycling process, thus a more representative specimen selection is possible.

For either measurement, single fiber tests show a great variation as expressed by Weibull's modulus. Higher values indicate lower variation and thus, table 3 shows that the juvenile fibers tend to show less variation. The Weibull modulus of the fibers recycled by microwave pyrolysis is by far the highest, leading to the assumption that additional imperfections are brought into the fiber. Nonetheless, when being interested in the influence of recycling to the fiber, single fiber testing is a valuable method. It can be faster than producing resin-impregnated specimen and testing composite specimen. More important, possible influences of the interface between fiber and matrix are precluded. Further, determination of the fiber appearance, for example SEM, can contribute to a well-founded understanding of fiber recycling and their properties.

Still, single fiber testing is challenging due to the variations between individual fibers, the small dimensions and low forces as well as the resulting high demands on the accuracy of the testing devices.

Therefore, the set up shown in figure 4 will be further developed. A laser diffractometer will be used to measure diameter and roundness of the fibers. Particularly for recycled fibers, a direct measurement of the diameter is preferable as the fiber's density is usually unknown for recycled material. Further, a method to determine the accurate elongation will be investigated.

6. Conclusion

Three sets of carbon fibers were investigated using single fiber testing methods and scanning electron microscopy. An addition, a new testing device has been set up and tested.

Using the newly built SiFiT device, it was possible to accurately measure tensile strengths of carbon fibers compared to those of the commercial provider. Further, the SiFiT was able to correctly measure with varying clamping lengths, low initial forces and variable cross head speeds, thus keeping a constant strain rate. However, several steps must be undertaken to fulfill the remaining goals listed in chapter 3.

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