RECYCLING OF LONG CARBON FIBERS, PART I: DEVELOPMENT OF A HIGH ALIGNED RCF-SLIVER FOR A BINDER TAPE MANUFACTURING PROCESS

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Abstract

For maximum utilization of carbon fiber properties and to use carbon fibers (CF) in a material saving but also load compatible way, it is necessary to process them with maximum orientation. For this purpose the recycled carbon fibers (rCF), which, to date, are mainly used in powder form or with less orientation in nonwovens, must be oriented and converted into semi-finished products similar to those made of primary CF (continuous tows, tapes). The presented process chain shows that it is possible to process rCF into a semi-finished product, the so called sliver, using textile processing technologies. Therefor the rCF are oriented by mechanical work with the aid of flexible clothings in a carding process and transferred in a linear shaped structure (sliver). The use of a three-cylinder drafting device has made it possible to further increase the orientation of the sliver. Measurement of the rCF orientation showed that the orientation could be increased by 20 % through a drafting system. To determine the orientation of the rCF the surface of the sliver was analysed with a camera under polyarized light. The results of the new technology were compared using a computer tomograph. Finally the possibilities of converting a high aligned rCF-sliver into a bindered sliver for further processing are shown.

1. Introduction

The global market for carbon fiber reinforced polymer (CFRP) grows constantly. Despite many advances in the composite production technologies, only 70 % of the applied carbon fibers (CF) end up in application, so 30 % of the fiber input become production waste [1]. Current recycling concepts include a utilization of the recycled CF (rCF) as short fiber reinforcement or in poorly oriented long staple semi-finished products which means a downcycling for the CF in both cases.

To make a better use of rCF the Deutsche Institute für Textil- und Faserforschung (DITF), Denkendorf, Germany together with the Institut für Verbundwerkstoffe GmbH (IVW), Kaiserslautern, Germany, Honda R&D Europe (Deutschland) GmbH, Offenbach, Germany and ELG Carbon Fibre Ltd., Coseley, United Kingdom carried out a project to develop a bindered sliver out of long rCF (LrCF) for thermoset matrix systems with a high alignment and parallelization of the LrCF (see Figure 1). Within this project all process steps, starting with CF recovery to the testing of the thermoset structures built out of the LrCF bindered sliver, were considered.

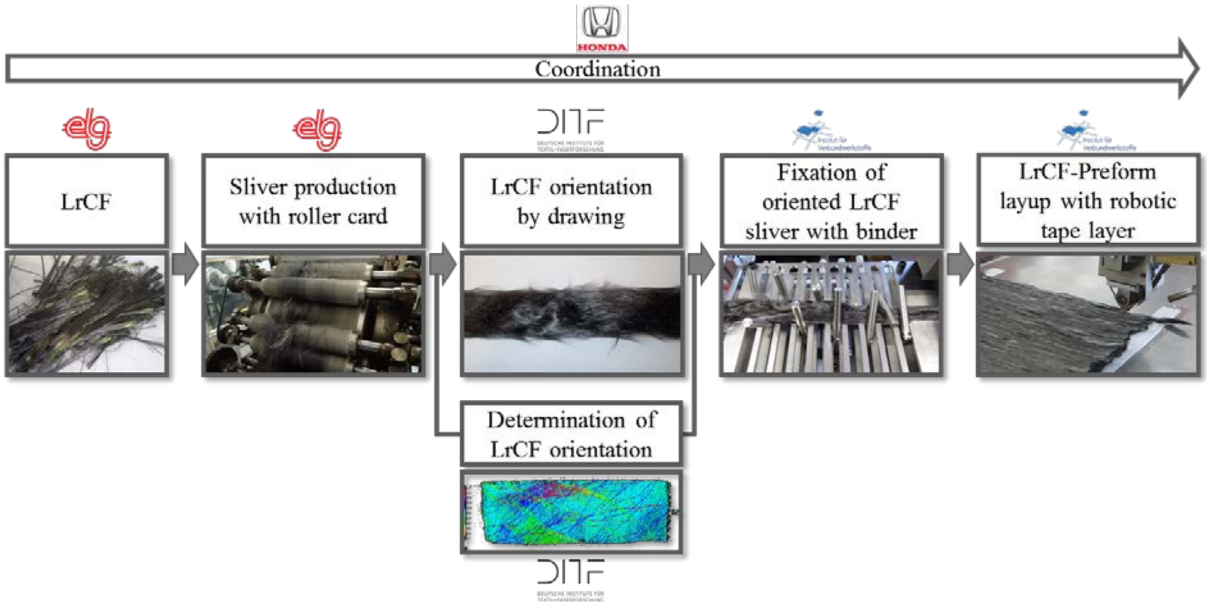

Figure 1. Schematical depiction of LrCF process chain starting from LrCF

2. Processing LrCF into a sliver

2.1. CF reclamation -

There are different processes for recycling CFRP, depending on which matrix material was used for the CFRP, whether the fibers and the matrix are to be recovered and what CF length is to be retained at the end of the recovery. In CF recovery, the matrix material is usually removed by solvolysis or pyrolysis. Solvolysis is currently only practicable for special matrix systems that can be separated again with solvents after consolidation or for pure CFRP waste for which suitable solvents are available. This is currently restricting the use of solvolysis. In most cases, the fibers are therefore thermally separated from the matrix by pyrolysis [2]. For this purpose, the CFRP are chopped and heated to 400 °C to 1000 °C under inert gas atmosphere. At the end of the pyrolysis process a oxidative atmosphere has to be used to remove the remaining deposits The chopping that is necessary for the pyrolysis process breaks the CF so they are available as staple fibers at the end of the process. ELG Carbon Fibre uses the pyrolysis technology to recover CF from CFRP.

To ensure a uniform raw material over the whole development project ELG Carbon Fibre prepared 150 mm long rCF from repurposed dry fibres waste. In the next step the LrCF have been pyrolyzed and then processed. To guarantee that the oxidative atmosphere during the pyrolysis did not affect the strength of the carbon fiber a single fiber strength measurement was carried out. The single carbon fiber strength before and after the pyrolysis was measured with a Favimat from the company Textechno Herbert Stein GmbH & Co. KG, Moenchengladbach, Germany. The tensile strength and modulus shown in Table 1 are mean values from 30 measurements each. The mean value of the single fiber length is generated out of 150 single fiber measurements that have been performed by hand with the help of two tweezers and a ruler. It is very difficult to isolate a single CF without breaking the CF therefore the measured CF length strongly depends on the skills of the laboratory assistant.

Table 1. CF properties before and after pyrolysis

Property	Virgin CF	Pyrolysed CF
Tensile strength [GPa]	4.06	3.86
Tensile strength confidence interval (95 % range) [GPa]	± 1.0	± 1.3
Tensile modulus [GPa]	207.07	209.62
Tensile modulus confidence interval (95 % range) [GPa]	± 11.3	± 8.4
Mean CF length [mm]	93.2	90.3
CF length confidence interval (95 % range) [mm]	6.5	9.6

Table 1 shows, that the pyrolyse process does not affect the properties of the LrCF.
Pictures taken with a scanning electron microscope (SEM) (Figure 2) show that after the pyrolysis the carbon fibers have a clean surface.

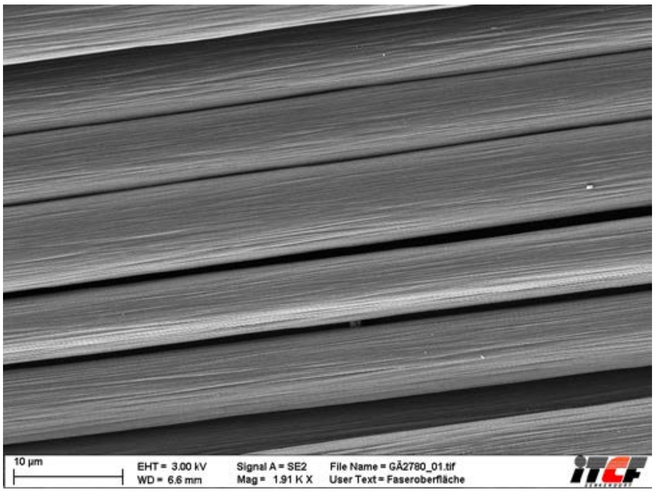

Figure 2. SEM picture of pyrolysed CF from the ITCF (part of the DITF)

The quality and fiber surface properties of the LrCF form the basis for all further processing. The LrCF properties like length or sizing have a significant effect on fiber fiber friction, fiber shortening, fiber waste and the transport stability of the sliver between the process steps.

2.2. Textile processing – carding process

After the CF have been recovered from CFRP scrap, they are available as unoriented tuft which have to be processed into an oriented semi-finished product in the next steps. Starting with the fiber opening and orientation on a carding machine (roller card), it was investigated which parameters result in a consistent sliver quality with a maximum of CF length. The roller card used by ELG Carbon Fibre consists of several rotating rollers (pairs of worker and stripper) with wires arranged around a large drum, the cylinder (see Figure 3). The LrCF run over the rotating cylinder, are repeatedly combed by the wires due to the relative speed between the cylinder and the rollers and are opened and oriented down to the individual fibers. At the end of the roller card is a cone which compresses the fibers and combines them to a sliver.

Figure 3. Exemplary a roller card with flexible wires processing LrCF of the DITF

The sliver produced by the roller card is buffered in cans and then fed into further processing (drafting system). Because of the necessary mechanical work that is applied to the LrCF fiber shortening occours. The aim of ELG Carbon Fibre was to reduce the fiber shortening as much as possible.

3. LrCF alignement

In the production of the sliver it is a technological necessity that the LrCF at the delivery of the roller card have a defined entanglement. This entanglement is necessary to hold the LrCF card web together over the working width of the roller card by fiber fiber friction. If the LrCF were only oriented in machine direction the card web would fall apart due to the lack of contact points at machine cross direction. To increase the preferred direction of LrCF generated by the roller card in the sliver (ideally, all LrCFs should be unidirectional), a drawing process is performed after the carding.

3.1. Sliver drafting system

Due to the draft of the card web which is formed to a sliver, the fibers within the sliver are stretched and aligned in the drawing direction by fiber fiber friction. In this way, the LrCF can be oriented in the processing direction as far as possible. For this purpose, the sliver is fed to a drafting system consisting of several pairs of rollers. The roller pairs have an increasing radial speed from intake to delivery. By clamping the sliver on the individual roller pairs, the differences in radial speeds between two clamping points apply a tensile force to the sliver, which is transmitted in the longitudinal direction of the sliver by fiber fiber friction. The described processes cause a draft and alignment of the fibers in the drawing process.

The type of the drafting system and the possible parameters for drawing the slivers depend on the fiber components used. The drawing parameters must be adapted to the fiber length distribution and the properties of the rCF (e.g. sizing mass on the rCF). In order to process the LrCF with an average fiber length of approx. 90 mm, two basic drawing technologies were investigated. The first technology was a chain gill draw frame, which is usually used for processing long staple fibers with fiber lengths greater than 80 mm. This technology was not effective for processing the LrCF, since the needle field, which generates a fiber guidance between the clamping points of the drafting unit, causes too much damage (shortening) to the LrCF.

The second technology with which it was possible to draw the LrCF sliver is based on a three-cylinder drafting system of a DITF laboratory spinning machine (see Figure 4). The draw frame consists of three cylinders and therefore, has two drawing fields. The first drawing field between cylinders 1 and 2 represents the pre-draft and shows a compaction of the sliver in the middle of the drawing field. The main-draft field between cylinders 2 and 3 does most of the work and has a rotating apron for better fiber guidance between the clamping points. Figure 4 shows beside the drafting system in closed state a LrCF sliver as it is processed on the draw frame.

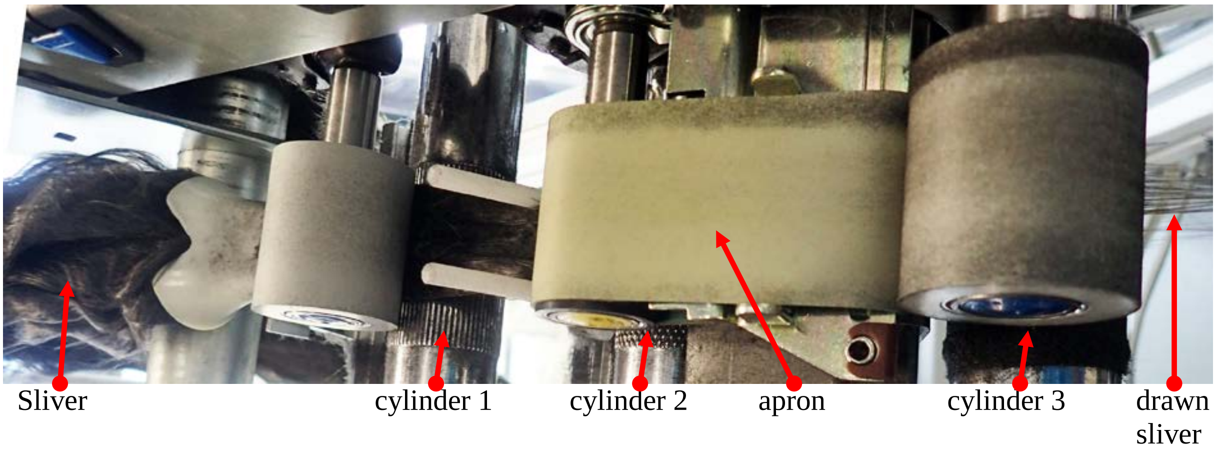

Figure 4. Three cylinder roller drafting device with LrCF sliver

3.2. Results of drawing process

The result of the optimization of the drafting system is a very well oriented sliver. Figure 5 shows the comparison between a sliver before and after drawing in the drafting system. Necessary adjustments at the roller draw frame device affected the pressure of the top rollers, which was reduced to 15 daN for all three pairs of rollers. The draw frame widths were set to 75 mm for processing the LrCF and a process speed of 10 m/min was achieved with a total draft of 2.5.

Figure 5. Left: rCF sliver from roller card, Right: High aligned rCF sliver after drawing

The fiber guidance of the smooth uncrimped LrCF with a large fiber length distribution poses a challenge for the drawing process. Any non-uniformity regarding mass or sizing of the LrCF will directly show irregularities in the sliver and degrade the bindered tape and the CFRP quality.
In general, the challenge of drawing the slivers is that fiber accumulations are formed from the short fiber content produced during rCF processing. The rCFs have high fiber fiber friction and must be guided and retained in a defined manner during the drawing process. If the guidance is inadequate or parameters are not selected optimally (e.g. draft is too high), the fiber components are segregated and the sliver uniformity degrades. The interaction of rCF properties and the draft in the drafting system is decisive for good fiber orientation. If, for example, the draft is selected too high, the LrCF will be disoriented due to incorrect draft behaviour in the drafting system (see Figure 6)

Figure 6. Poorly oriented sliver due to too high draft

4. LrCF orientation measurement

To verify the improved LrCF orientation after the carding and drawing process, a measurement method was investigated.

The basic idea of a camera that uses polarized light is that CF does influence polarization of impacted (infrared) light very strong. This material property uses a prototype camera technology called POLKA from the Fraunhofer-Institut für Integrierte Schaltungen (IIS), Erlangen, Germany. The principle of POLKA is that light has three relevant properties: wave length (color), intensity (brightness) and polarization (not realized by person or normal cameras). Light is polarized by filter, reflexion on non metal surfaces and by diffusion. The light polarized by the sample can be detected with the POLKA camera which can detect polarization by camera image sensor for each pixel. The POLKA system is optimal for analyzing fiber orientation of CF and delivers live images of orientation angle of CF with frame rates up to 200 frames/s.

In order to be able to trace the measurements of the POLKA camera system, samples were additionally examined with a computer tomograph model nanotom m from the company General Electric Sensing & Inspection Technologies GmbH, Frankfurt, Germany. The µ computer tomograph nanotom m is perfect suitable to examine textile- und plastic based materials. The fiber orientation could be determined by high-resolution CT scans showing the individual CF in combination with CT analysis software VGStudioMax.

4.1. Results of LrCF orientation measurement

The research work showed that a polarization camera together with the backup of computer tomography scans could pose a good means to exam the three dimensional LrCF orientation (Figure 7).

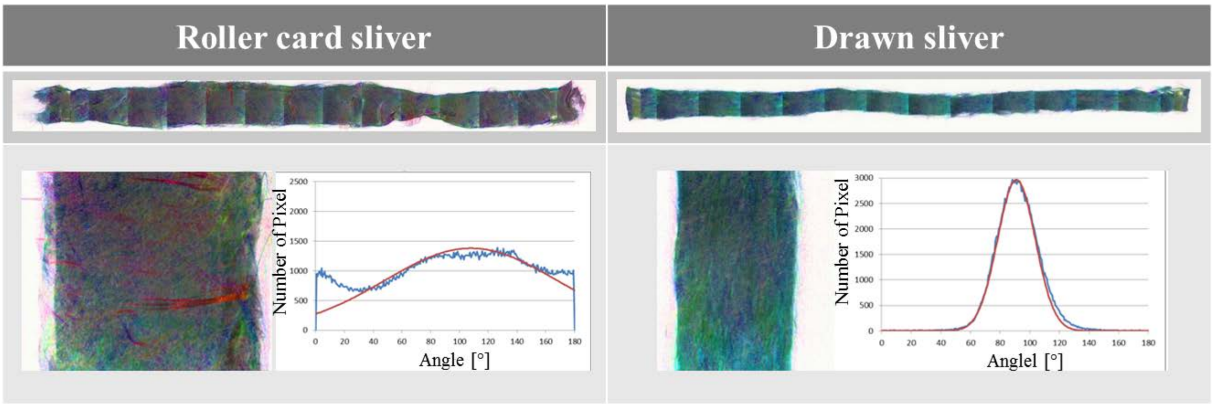

Figure 7. LrCF orientation before and after drawing measured with a polarization camera

Figure 7 shows that the results made during the tests to improve the fiber orientation by drawing the slivers can be measured. The measurement (Figure 7) shows the enormous increase in fiber orientation from the sliver produced by the roller card to the sliver after the draw frame (increase of orientation is approx. 20 %). Due to the measuring principle, the maximum fiber orientation in the longitudinal direction is not at 0° as usual but at 90°. The 90° corresponds to the longitudinal orientation of the sliver, since the measuring device works at right angles to the longitudinal axis.

Both measurment methods (POLKA and CT) represent the change of orientation by drawing the slivers with the three cylinder draw frame. The CT scans of the samples show comparable results to the POLKA method in distribution of fiber angles shown in histograms. In general the CT scan is more precise, as it analyses the fiber distribution over the complete scanned volume instead of a surface area. But the CT scan can only be done for a small part in required high resolution, which is possibly not representative (Figure 8). The POLKA images show the same orientation structure like in CT surface view and the scanned region is tremendous bigger. The result of the comparison of the both methods is that the POLKA image of a small piece is not reliable, but the POLKA information of a 100 x 9 cm² sample is more significant than a CT measurement of a 2,5 x 1 cm² sample.

Figure 8. CT scan (left) 2,5 x 1 cm² compared to POLKA measurement 100 x 9 cm² (right)

Final tests showed that the fiber orientation can be best detected on the tape surface (after binder application to the sliver at the IVW) because the tape has a smaller volume compared to the sliver and therefore more LrCF can be detected at the same depth of field. To use this effect, the sliver was covered and compressed with a transparent plastic plate for measurement. For measurement with the CT the sliver was compressed using a rubber hose. The measurements also showed that the Polka camera does not detect the orientation of the binder material because it is made out of thermoplastic material.

5. Further processing of high aligned LrCF sliver

The highly oriented LrCF sliver out of the drafting process must be stabilized for winding and further processing. Here, the sizing of the LrCF fibers plays an essential role. To stabilize the oriented LrCF slivers they were processed at a binder application rig from the IVW which will be presented in a subsequent presentation. The whole process chain is shown in Figure 1.

6. Conclusion

The presented devolpment shows, that the orientation of LrCF by means of textile processing is possible. The combination of a carding process with a subsequent drawing process increases the LrCF orientation wthin a sliver tremendously. Furthermore the sliver is a semi-finished product which can be further processed into a tape for a tape laying process like it is performed with new CF tapes.

During the development the field of tension resulting from good sliver adhesion due to high fiber fiber friction and the highest possible CF orientation reached in the drawing process by sliding effects of the LrCF (low fiber fiber friction) was challenging. The trials in this field show the importance of the LrCF quality including the sizing and the fiber length and the need to optimize the parameters of each process step to the fiber quality from the fiber recovery to the tape laying process to achieve the best CFRP properties.

In general it can be statet that CF are being shortened during treatment but there is no fiber degradation through pyrolysis if it is operated under the right conditions. Drawing the sliver increases intense the fiber orientation but the drawing has to be performed with a suitable draw frame. The measurement of fiber orientation with the help of a POLKA camera provides reliable information, especially for sample sizes in the range of 100 x 9 cm².

Further processing of the drawn sliver in combination with possible CFRP performance out of tapes made of the drawn sliver is presented in a subsequent presentation of the Institut für Verbundwerkstoffe GmbH (IVW), Kaiserslautern, Germany.

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