Development of heavy tows from recycled carbon fibers for low-cost and high performance thermoset composites (rCF heavy tows)

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Introduction

Carbon fiber-reinforced plastics (CFRP) are increasingly used in lightweight applications due to their high stiffness and strength as well as low density, especially in aerospace, transportation, wind energy, sports equipment or construction. Global demand of CFRP is predicted to increase to 197,000 t/a [1] by 2024, almost tripling compared to 2011. This shows an urgent need for [2, 3] solutions to recycle the high quality carbon fiber (rCF) in terms of the circular economy. This is necessary not only due to strict legal regulations, but also for ecological and economic reasons. In recent years, numerous research institutes and companies developed solutions for the reuse of rCF in the fields of nonwovens [4-6], injection molding [7, 8] or as hybrid yarns [9-10]. However, the majority of these works involve the use of rCF in combination with thermoplastic fibers for thermoplastic composites. In the field of rCF-based thermoset CFRP, mainly rCF nonwovens made of 100% rCF have been so far developed [4-6]. Since the fibers in the nonwovens mostly have a limited length and a low orientation and process-related additional high fiber damage occurs, with these materials only maximum 30% of the composite characteristic values of CFRP components made of carbon filament yarns can be so far achieved.

Currently, the matrix systems used in the field of high mechanical loaded CFRPs are predominantly thermoset. Such components exhibit high dimensional stability, high stiffness and strength as well as are suitable for the implementation of complex component geometries due to low-viscosity matrix systems. However, primary carbon filament yarns are particularly used for these components due to the insufficient properties of rCF. In addition to low sustainability, the utilization of these filament yarns result in at least 200 % higher cost [11-13]. The production of primary carbon filament yarn requires a high energy demand of about 230 MJ/kg [14, 15] with a CO_2 emission equivalent to 20 kg CO_2/kg CF [16, 17]. Here, a significant improvement of the CO_2 balance is required to make a substantial contribution to the envisaged climate protection goals of the Federal Republic of Germany [18, 19] and the EU [20]. For this reason, the focus of the project work is the development of novel, sustainable rCF heavy tows made of recycled carbon fibers (rCF) and associated manufacturing technologies for the implementation of cost-effective thermoset composites with high mechanical performance.

Development of a process chain for the production of rCF heavy tows at ITM

The developed process chain for the production of rCF heavy tows with high mechanical performance for complex shaped thermoset composites is schematically shown in Fig. 1.

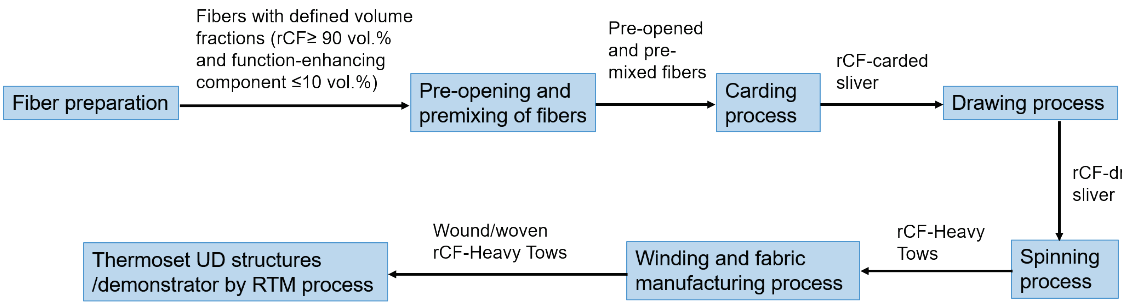


Fig. 1: Schematic diagram of the process chain for the development of rCF-Heavy Tows

The properties of the selected recycled carbon fibers (rCF) and hotmelt staple fibers copolyamide (SKF, EMS-Griltech GmbH) are summarized in Tab. 1.

Tab. 1: Fiber properties

Fibers	Mean fiber length (mm)	Mean fiber diameter (µm)	Breaking elongation (%)	Young's modulus (GPa)	Mean tensile strength (MPa)	Surface energy (mN/m)	
						Dispers	Polar
rCF	80 ± 2; 100 ± 2	5.5 ± 0.10	1.7	268 ± 59.6	3558 ± 758	14.1	9.1
SKF	100 ± 2	34.5 ± 0.6	147.9	0.30 ± 0.1	268 ± 20	14.6	8.3

The selected and characterized rCF reinforcing fibers as well as the hotmelt staple fibers copolyamide (SKF) as function enhancing fibers for imparting adhesion between the rCF were prepared in the first step for the fiber opening with defined fiber volume fractions, average fiber length and properties to meet the requirements of the process chain and the composite properties. For this purpose, the pre-opening and pre-mixing device of the fiber preparation plant for gentle processing of rCF and functional fibers was optimized with respect to the speed and distance of the opening unit. Using the modified pre-opener and pre-mixer, rCF and SKF with defined volume fractions (rCF/SKF: 98/2 vol.- %, 95/5 vol.- %, 90/10 vol.- % and 100/0 vol.- %) were gently and uniformly pre-opened or additionally pre-mixed.

The pre-opened and pre-mixed fibers were then processed into a sliver in the carding process. Main challenges of the carding process were the realization of carded slivers with high uniformity and low fiber damage of the smooth, very fine and extremely lateral-force sensitive CF. For this purpose, technological modifications were designed and implemented to the carding technology (e.g. to reduce the intensity, with which the stripper-worker roller pairs penetrate the fibrous web) and components were further or partly newly developed (e.g. tangential feeding of fibers to the breaking cylinder) for gentle fiber guidance without damaging deflections (deflection radius at least 8 mm). In addition, small amounts of sizing or, finishing agents (≤ 2.5% by volume) were applied to the rCF for optimum friction between the fibers as well as between fibers and roll clothing. Optimum machine and textile technological parameters were also determined on the modified carding line and thus carded slivers were produced from pre-opened and pre-blended fibers with defined fiber volume fractions of rCF and SKF and these were characterized. The card slivers are thus also available for investigating the drawing process.

The development and implementation of a drawing process was another part of the project. The objectives were to increase sliver uniformity and reduce fiber damage, especially at the auto-leveling unit of a draw frame. To this end, the following approaches were pursued.

According to approach 1, the scanning roll load and surface of the draw frame's auto-leveling unit were adapted to the extreme transverse force sensitivity of the rCF-fibers. Approach 2 pursues the further development of the process control by multiple drafting and increased number of doubling. In addition, the standard fluted bottom rolls of the drafting system were replaced by topocrom-coated bottom rolls with optimized surface properties and high wear resistance. After completion of the modifications, the relevant drafting system-related parameters, such as roller spacing, top roller load, amount of draft, auto-leveling as a function of fiber lengths and sliver non-uniformities, were investigated and the best possible parameters for the production of draw frame slivers were determined, manufactured and characterized. The draw frame tapes are available for the development of rCF heavy tow.

The focus of the research work was the development of novel rCF heavy tows and the associated manufacturing technologies. For this purpose, a modular test set up was designed and implemented. This consists, among other things, of a drafting system for supplying the drawn sliver with defined sliver count, a thermostabilization module for increasing the drawn sliver strength by melting added hotmelt adhesive fibers in it, a guiding nozzle for compacting the bonded the fiber structure into rCF heavy tows, and a winding module (Fig. 2). With the help of the test set-up, existing textile-technological interactions and technology limits were investigated in depth. To derive best possible manufacturing parameters, rCF heavy tows were produced from rCF (≥90 vol. %) and small amounts of hot melt adhesive fibers (copolyamide SKF 2/5/10 vol. %) by varying the fineness, the intensity of heat input of thermo-stabilization unit, the diameter of the guiding nozzle and the production speed. Using the best possible production parameters, rCF heavy tows of different finenesses were produced and the fineness-related stress-strain behavior (DIN EN ISO 13934-1) as well as fiber lengths were determined by means of image processing method developed at ITM. The rCF heavy tows are available for the investigation of further processability for fabric structure by means of weaving technology and for the production of thermoset composite panels by means of RTM (Resin Transfer Molding) processes. For composite tensile tests, unidirectional (UD) composite sheets were manufactured and characterized according to DIN EN ISO 527-5/A/2.

The manufactured fiber slivers, rCF heavy tows and UD test sheets with defined fiber volume fractions are shown in Tab. 2.

Tab. 2: Exemplary selected products with defined fiber volume fractions

Fiber volume fraction [Vol.-%]		Carded sliver (KB)	Drawn sliver (SB)	rCF-Heavy Tows (HT)	Unidirectional (UD) composite slab
rCF	SKF				
90	10	KB1-rCF/SKF-90/10	SB1-rCF/SKF-90/10	HT1-rCF/SKF-90/10	UD1-rCF/SKF-90/10
95	5	KB2-rCF/SKF-95/5	SB2-rCF/SKF-95/5	HT2-rCF/SKF-95/5	UD2-rCF/SKF-95/5
98	2	KB3-rCF/SKF-98/2	SB3-rCF/SKF-98/2	HT3-rCF/SKF-98/2	UD3-rCF/SKF-98/2
100	0	KB4-rCF-100	SB4-rCF-100	HT4-rCF-100	UD4-rCF-100

rCF: recycled CF
SKF: Co-polyamide/hot melt adhesive fibers

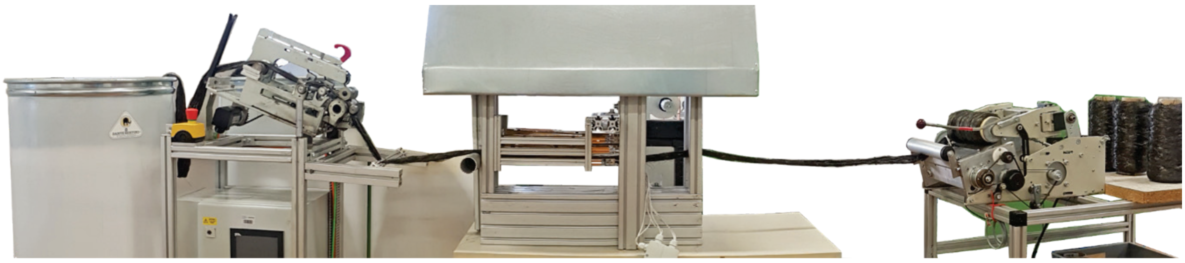
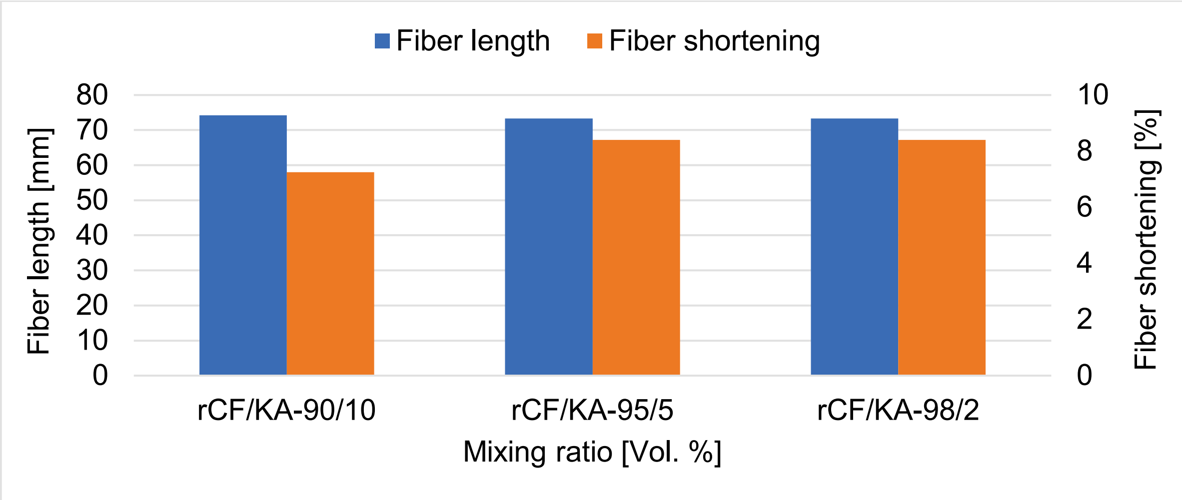


Fig. 2: Modular test set-up for processing of twist-free rCF-Heavy Tows from drawn sliver

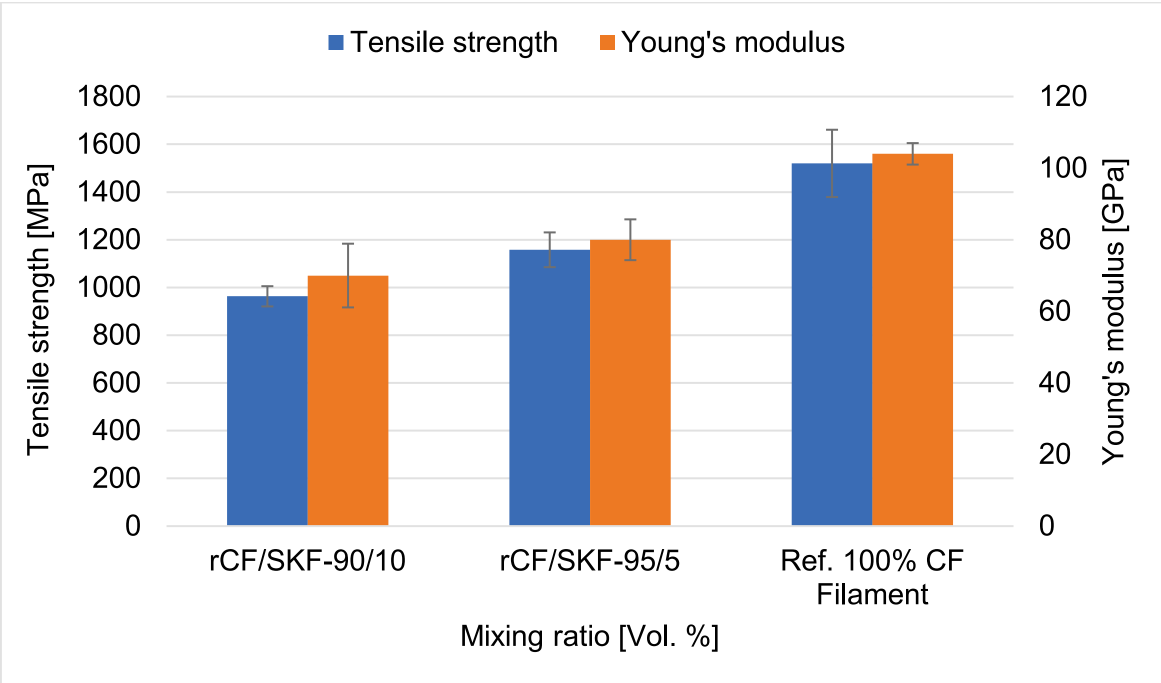
Results (selected)

The technical-technological investigations and developments of test set-up within the scope of this work allow the gentle processing of smooth, very fine and extremely cross-force sensitive rCF to rCF heavy tows with very low fiber damage and high fiber orientation and resultant composites with high mechanical performance. By optimizing the technological and machine parameters of the carding line, fiber damage or fiber shortening was reduced from ca. 80% to less than 10%. Sliver uniformity was significantly improved by optimizing the draw frame.

The influence of the different proportions of hotmelt fibers on fiber damage or fiber shortening during the processing of rCF in the carding process and on the mechanical properties of the composite slab based on the rCF heavy tows is shown as an example in Fig. 3. The fiber damage or fiber shortening in the carding process decreases slightly with increasing proportion of hotmelt fibers. The function-enhancing component, e.g. hotmelt adhesive fibers, leads to a cohesive bond between the fibers and reduces the mechanical stress on the rCF caused by the card clothing. As a result, damage to the rCF during the carding process is reduced. The tensile strength and Young's modulus of composites are measured 1158±72 MPa and 80±5.7 GPa and 963±42.4 MPa and 70±8.9 GPa, respectively, when the hotmelt fiber content is 5 and 10% by volume. These values correspond to about 77% of the tensile strength and Young's modulus, respectively in comparison to the composites based on carbon filament yarns. Even a low proportion of approx. 5 vol.-% hotmelt fibers results in uniform, trouble free and gentle processing of rCF, which leads to uniform fiber structures and composite structures. A further increase in the proportion of hotmelt fibers to 10 vol.-% can still produce uniform fiber structures and composite structures, but here the proportion of hotmelt fibers is so large that it has a negative effect on the composite properties due to the lower fiber volume ratio.



(a)



(b)

Fig. 3: rCF-Heavy Tows - Influence of rCF volume fraction on (a) average fiber length or fiber shortening in the carded slivers and (b) tensile strength of thermoset composites (fiber volume ratio 50 vol.-%).

Summary

Within the framework of the IGF research project (21612 BR), the entire process chain for the industrial production of novel twist-free rCF heavy tows was developed at ITM. In particular, a novel technology for the production of rCF heavy tows based on recycled carbon (rCF ≥ 90 vol.-%) and hot melt adhesive fibers (< 10 vol.%) was designed, constructed and successfully implemented. This includes fiber preparation, the carding process for card sliver formation, the stretching process for drawn sliver formation, and the final fabrication of the rCF heavy tows from rCF and hot melt adhesive fibers in a newly developed test set-up. The suitability of the developed technology is demonstrated by the implementation of rCF heavy tows with different rCF types, fiber lengths and fiber volume contents and a demonstrator. Fig. 4 shows the process flow developed at ITM from rCF to demonstrator. The developed rCF heavy tows with finenesses between 3000-7000 tex and their further processability into textile semi-finished

products were successfully demonstrated. The developed rCF Heavy Tows and composites based on them exhibit a maximum composite tensile strength and a maximum Young's modulus of 1158±72 MPa and 80±5.7 GPa, respectively. The rCF Heavy Tows are thus applicable for low-cost thermoset composites with high performance and complex geometry. Thus, the developed rCF Heavy Tows offer a very high innovation and market potential in the fields of materials and materials, lightweight construction, environmental and sustainability research, and resource efficiency. This opens up the opportunity for SMEs in the textile industry to develop new products and technologies for the fiber composite market and to establish themselves as suppliers for the automotive, mechanical engineering and aerospace, medical and sports equipment industries.

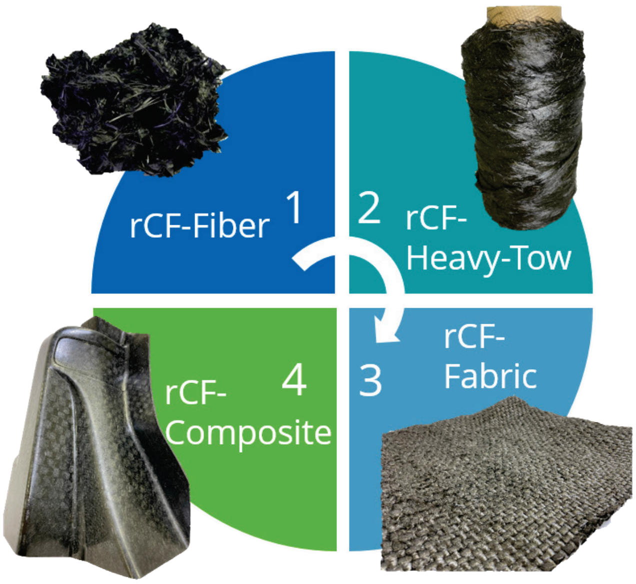


Fig. 4: Material flow of the process chain: from fiber to rCF heavy tow, fabric to composites

Acknowledgments

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