NONWOVENS MADE OF RECYCLED CARBON FIBRES (rCF) USED FOR PRODUCTION OF SOPHISTICATED CARBON FIBRE-REINFORCED PLASTICS

Katharina Heilos[1], Holger Fischer[2], Marcel Hofmann[1] and Andrea Miene[2]

[1]*Sächsisches Textilforschungsinstitut e.V. STFI, Chemnitz (DE)*
[2]*Faserinstitut Bremen e.V. FIBRE, Bremen (DE)*
Katharina.Heilos@stfi.de; Fischer@faserinstitut.de

Abstract: *A qualified value-added chain for the re-use of recycled carbon fibres (rCF) as nonwovens in sophisticated fibre-reinforced plastics has been set up within the research project RecyCarb. The progress in processing technology was accompanied by setting up a reliable scheme of quality assurance. This article focuses (1) on the use of different web formation and bonding technologies to produce nonwovens (carding / airlay), ranging from highly-oriented to quasi-isotropic nonwoven structures, and (2) on the achievable mechanical properties of fibre-reinforced plastics produced using different processes from a range of typical rCF lots. Press-moulded composites reached tensile strength of approx. 530 MPa (CD) & 330 MPa (MD), and Young's moduli of 50 GPa (CD) and 31 GPa (MD). Similar values were obtained by resin-transfer moulding (RTM). Prepreg-based composites displayed tensile strength up to 330 MPa and Young's modulus up to 29 GPa. In addition, first experiments showed that repeated recycling of carbon fibres is possible with minor loss of stability.*

Keywords: *recycled carbon fibres (rCF); nonwoven; carbon fibre-reinforced plastics (CFRP); lightweight construction; mechanical properties.*

1 INTRODUCTION

The market predictions with an increase of 10% p.a. for carbon fibre-reinforced plastics (CFRP) are excellent 1. Consequently, there are increasing amounts of production scraps as well as materials reaching the 'end-of-life'. Actually, these materials can be recycled by milling or shredding and subsequent pyrolysis. The resulting products are entering the growing markets for recycled carbon fibres (rCF), although they are inhomogeneous in their morphology 2. These mixtures comprise randomly oriented roving residues & filaments in wide fibre length distribution. A few years ago, the only established industrial market was use of short fibres or milled material for injection moulding - a product more improving the antistatic properties than the mechanical properties of plastics. In addition, at that time there were no definitions for long rCF (>10 mm) and final products concerning necessary qualities (length distribution, minimum tenacity, homogeneity, etc.), sampling in process and quality of the final products compared to those made of virgin fibres 3.

Beside short fibre reinforcement in injection moulding 4, sophisticated applications of rCF in composites have been developed, like thermoplastic hybrid yarns 5, so-called 'secondary rovings' made of rCF plus binder 6 or nonwovens made either of pure rCF 7, or as hybrid nonwoven in combination with e.g. flax 8, 9.

Meanwhile basic principles for characterisation of rCF have been defined, enabling the quality classification for rCF off-line in the laboratory 10.

Another aspect is the environmental issue of using carbon fibres. From the energy demand of carbon fibre production and the high price of virgin fibres it is clear, that recycling and re-use of the fibres should be the favourable way. An actual review of LCA studies shows, that there are environmental advantages in the cradle-to-gate phase of rCF reinforced composites for specific cases, but as well the future demand for more intensive LCA studies 11.

Result of preliminary work was processing of long, but not endless recycled carbon fibres by means of the carding principle, using either 100% carbon fibres or blends with natural fibres and/or synthetic fibres first-time in pilot plant scale 12. The results depicted clearly, that web formation is possible not only from 100% primary carbon fibres, but as well from 100% recycled carbon fibres via mechanical carding 3, 12. The resulting average fibre length was approx. 85% of the pre-cut. These carbon fibre nonwovens were sufficient in strength, making them suitable as semi-finished products for CFRP-structures. Furthermore, a successful approach to produce composites from rCF preforms 13 has been reported.

Subsequently, within the frame of the research project RecyCarb 14 the research focus was set on:

- Process scale-up for nonwoven production into industrial and economical relevant scale with respect to the quality requirement,
- Set-up of a process-integrated monitoring of quality parameters,
- Evaluation of the effects of different nonwoven technologies, including first-time application of a combined nonwoven process for generating quasi-isotropic nonwoven structures.

Aim of the project work was to set-up a qualified value-added chain for recycled carbon fibres (rCF) by closing the technological gap between rCF und functional high-value re-use 14. The work comprised the definition of necessary initial quality & standards for consistent sampling in the process, process-attached monitoring in terms of Industry 4.0, reproducible intermediate quality by optimisation of the carding process and finally upcycling by re-use of rCF into high-value parts 15. The final report is available in German 14.

Consequently, meanwhile first rCF nonwovens are entering the commercial market, offered by several companies 16, 17.

This article highlights the achievable quality levels for rCFRP made of rCF nonwovens using different production methods suitable for industrial use.

2 MATERIALS & METHODS

2.1 Materials

Three types of rCF are used: off-cuts (mainly non-crimp fabrics) provided by partners of the consortium (Schmuhl Faserverbundtechnik GmbH, Liebschütz, DE and TENOWO GmbH, Hof, DE), hoover waste of the non-crimp fabric production (Saertex GmbH, Saerbeck, DE) or reclaimed fibres by pyrolysis, i.e. EoL-CFRP supplied by Eissmann Cotesa GmbH, Mittweida, DE and pyrolysed rCF by CarboNXT GmbH, Wischhafen, DE.

Offcuts and hoover waste were selected as suitable starting materials for nonwoven production due to their comparable virgin state concerning original sizing and resin free surface. Caused by the examined impregnation methods, the selected offcuts and hoover waste were appropriate for thermoset resins.

The pyrolysed rCF were supplied without sizing as a result of the pyrolysis, which has to be considered for the nonwoven production and the mechanical performance (fibre-matrix interface) of the resulting composites.

The fibre qualities have been analysed for each processed lot according to the scheme given below in section 2.4. Details of the results have already been published 18, with all production scraps in the range of 4,500 – 6,000 MPa tensile strength (exceptions not used in this work) and fibre diameter of 7 µm. Due to the demands of material input for the line, similar lots had to be mixed before processing. For this reason, the fibre strength in the produced nonwovens can be assumed to be 5,200±500 MPa; detailed values are not mentioned in the results. The pyrolysed fibres have been found to be approx. 10% lower in tensile strength and Young's modulus 10.

2.2 Nonwovens production

Nonwovens were produced on a dust-proof pilot plant with 1 m working width at the Center of Textile Lightweight Engineering at STFI, Chemnitz, DE, described in 15, 19. An additional exhaust system enables a dust-reduced production. First the rCF material is cut to an average length of 50-120 mm using a guillotine cutting machine. This is followed by fiber opening and separation in a modified tearing machine. For the subsequent web forming the line offers two options: either airlay- or carding process.

In the aerodynamic web forming process (Airlay Card K12-direct from Autefa Solutions Germany GmbH, Friedberg, DE) the carbon fibers are transported via air-stream and are deposited randomly on a filter belt. Thus, the resulting nonwoven structure is quasi-isotropic.

Carding is performed by using a MiniCard unit, combined with a cross-lapper type Topliner, both supplied by Autefa Solutions Germany GmbH, Friedberg. In the carding process the carbon fibers are separated via a tambour (main cylinder) and several stripper/worker pairs, and are finally deposited in a zigzag pattern by the cross-lapper according to the requested mass per unit area. As result, anisotropic nonwovens are obtained, displaying higher strength in cross-direction (CD) than in machine direction (MD).

Finally, mechanical bonding is performed by needling or stitch-bonding process in order to produce the desired nonwoven. In needle-punching process barbed needles punch vertically in and out of the material, while in the stitch-bonding process an additional warp knitting thread is introduced to bond the nonwoven 20.

In addition, nonwovens were produced at TENOWO GmbH by using a carding/cross-lapping process combined with stitch-bonding.

Nonwovens were preferably produced with mass per unit area 300 g/m² in airlay process as well as in carding process. In addition, the possibility of a second recycling by using own nonwoven production scraps and using them again for producing rCF nonwovens has been examined. This resulted in so-called re-recycled nonwovens (rrCF nonwovens) with mass per unit area ranging from 100 to 250 g/m².

2.3 *Composite production*

The manufactured rCF-nonwovens were processed into so-called rCFRP by three different types of composite manufacturing processes: (1) hand lay-up by impregnation with epoxy resin, followed by compression-moulding to a thickness of 2 mm using a hydraulic column downstroke press at STFI, (2) resin transfer moulding by vacuum impregnation a Schmuhl Faserverbundtechnik GmbH, Liebschütz, DE and (3) the wet lay-up method with autoclave at Eissmann Cotesa GmbH, Mittweida, DE.

2.4 *Scheme of sampling & analysis*

A scheme for sampling has been developed for incoming rCF lots as described in 15, derived from DIN EN 12751:1999 21.

Incoming fibre lots have been characterised using Dia-Stron single-element analysis (Dia-Stron Ltd., Andover, UK) to assess fibre tenacity and Young's modulus acc. to DIN EN ISO 5079:1996 22, clamping length 3.2 mm. Preceding the tensile tests the cross-section of each single specimen was measured via laser beam. Up to 45 specimens were measured to ensure statistically firm results as described in 23. A part of the lots has been examined by SEM (Cam Scan CS24, EO Elektronen-Optik-Service GmbH, Dortmund, DE with Software analySIS 3.2, SIS Soft Imaging System GmbH, Münster, DE) to identify contaminations, dust, and/or fibre damages. Fibre length distribution of rCF lots shorter than 100 mm was analysed by image analysis FibreShape V6.1.2f with addon FiVer (IST AG, Vilters, CH), based on 2,500 up to >6,000 fibres, depending on sample homogeneity. The adaption of the sample preparation and measurement parameters has been described in 14. This enables the reproducible analysis of filter dust as well as inhomogeneous cut fibers.

If the incoming lots consist of roving snippets, the snippet length distribution was analyzed using a flat-bed scanner at 100 dpi and software FibreShape V5 as described in 10.

For the process control each three positions have been defined for off-line sampling as well as for on-line control via camera. Off-line sampling is scheduled for incoming lots control (fibre quality analysis), for the carded- or airlay-web and finally the needle felt (analysis of grammage). On-line control is scheduled at the airlay- and card supply (identification of contaminations), directly after card or airlay (control of fiber orientation and web homogeneity) and finally after needle-punching (control of fiber orientation and nonwovens homogeneity). Using this scheme enabled analytical control of the entire process and successful outcome of the processing experiments 15.

The system for on-line analysis developed within the project is based on CCD-camera technology and image analysis software, which has been developed at FIBRE, Bremen; DE. The analysis of the MD/CD ratio is conducted by means of the filament orientation distribution and displayed as histogram (detailed description in 14).

The manufactured laminates were analyzed in terms of tenacity according to DIN EN ISO 527-4/1b/2 24 and flexural strength according to DIN EN ISO 14125 25. The fibre volume fraction (FVF) has either been analysed acc. to DIN EN ISO 1172 26 or been calculated from nonwoven grammage, dimensions and weight of composite specimen.

3 RESULTS & DISCUSSION

Composite boards have been produced from the different types of rCF nonwovens mentioned above to examine the potential of these materials for lightweight construction applications. The results are presented separately for different typical production processes in the following subsections.

3.1 *Composites made of different types of rCF nonwovens using hand lay-up*

Two types of rCF have been used: pyrolysed fibres as well as production scraps, cut from dry non-crimp fabrics, hoover waste or rovings in different nominal length grades ('cut size').

The nominal and real fibre parameters as well as the nonwoven production parameters are listed in Table 1. These nonwovens were stacked to reach a grammage of 1.800 g/m² (6 or 9 layers, resp.), impregnated (hand lay-up) with epoxy resin and finally press-moulded to a thickness of 2 mm as described in section 2.

Table 1 Fibre and processing parameters

Sample name	**rCF properties**		
	nominal length [mm]	***roving length [mm]***	***nonwoven production***
pyrolysed fibre	60	45.5 ±24.7 after opener	carding, needle-punching 200 g/m² at STFI
50% pyrolysed fibre/50% scraps	60/100	n/a	carding, needle-punching 300 g/m² at STFI
scraps	50	36.5 ±12.3 after opener	carding, stitch-bonding 200 g/m² at TENOWO
scraps	70	45.6 ±15.4 after opener	
scraps	100	61.4 ±26.0 after opener	

As a result of the needle-punching, the boards contain a remarkable share of fibres in z-orientation. Thus, the boards did not comply exactly with the thickness of 2 mm. Therefore, the fiber volume fraction (FVF) was calculated individually from nonwoven grammage, dimensions and weight of specimen. It is listed in Table 2. From these data the average FVF can be given as 36±2%. Consequently, variations of grammage and pores have to be considered regarding FVF and mechanical properties of the boards, summing up to ±6%.

Sample specimens were cut from the resulting composite boards in machine- and cross-direction (MD and CD) to analyse their mechanical stability.

Table 2 Fibre volume fraction of the composites made from different types of rCF nonwovens

Sample name	Fiber volume fraction [%]
Pyrolysed fibre	34.2
50% pyrol. fibre/50% scraps	35.6
Scraps “50 mm”	37.9
Scraps “70 mm”	37.6
Scraps “100 mm”	35.5

The tensile strength values are presented in Figure 1. The samples were produced on two different lines: pyrolysed fibre and blends of pyrolysed fibres/scrap at STFI and scraps at TENOWO (cf. Table 1). Nevertheless, all samples display a predominant tensile strength in CD caused by the process set-up: the carded web with fibres oriented predominantly in MD is fed into the cross-lapper, where the orientation changes for almost 90°, giving a predominant CD orientation in the resulting nonwoven. Due to the different production equipment this effect is a bit more distinct for the pyrolysed fibre samples. It is easy to observe, that all composites produced from rCF scraps display the same tensile strength: 225-241 MPa in MD and 280-290 MPa in CD.

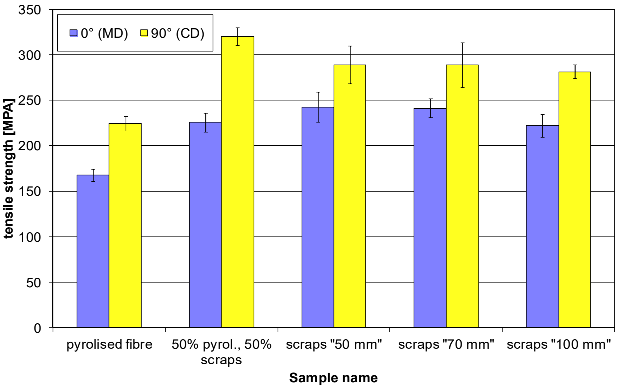

Figure 1 Tensile strength of the composites produced from different types of rCF nonwovens

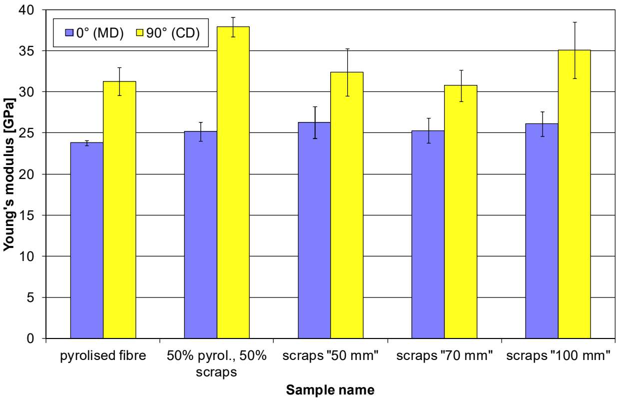

Figure 2 Young’s modulus of the composites produced from different types of rCF nonwovens

The differences between those three samples are within the limits of uncertainty. The sample made of 50% pyrolysed / 50% scrap fibres displays almost the same tensile strength in MD (225 MPa), and slightly larger tensile strength in CD: 320 MPa. This difference may be caused by the different number of web layers in the stack (6 instead of 9), which means as well a lower haul-off speed in the cross-lapper. This was found to cause a higher orientation in CD in previous research 19. The only sample with significantly lower tensile strength in MD (168 MPa) and CD (224 MPa) is the one made of 100% pyrolysed fibres. These fibres were more brittle than the scrap fibres. This caused not only higher losses in the nonwovens production (>45%, compared to 20-30% for scraps), but as well a shorter fibre length in the nonwoven. Consequently, all five samples are very similar in Young’s modulus, as displayed in Figure 2. The differences between the samples are within the limits of uncertainty (24-26 MPa in MD / 31-35 MPa in CD).

The flexural strength of the samples is shown in Figure 3. The same tendencies as for the tensile strength can be observed here.

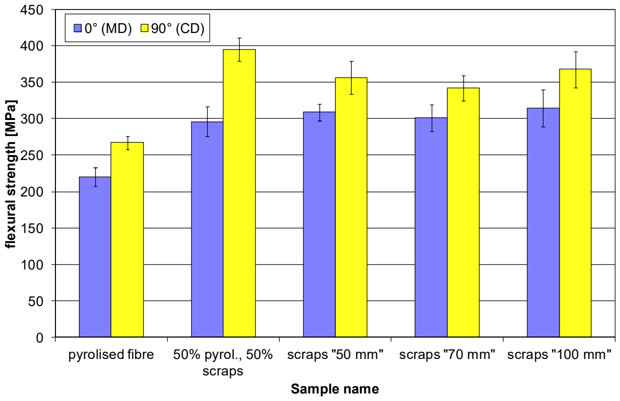

Figure 3 Flexural strength of the composites produced from different types of rCF nonwovens

Concerning the flexural modulus of the samples displayed in Figure 4 there is only one difference compared to Young's modulus: the samples containing pyrolysed fibres exhibit a higher level with 20 MPa (MD) / 25 MPa (CD) than the scrap samples with 15 MPa (MD) / 20-22 MPa (CD).

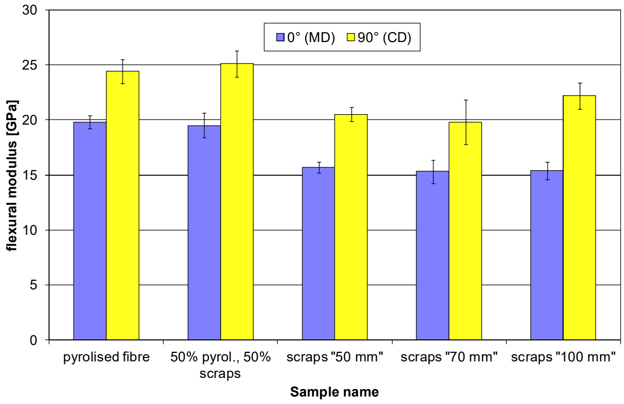

Figure 4 Flexural modulus of the composites produced from different types of rCF nonwovens

In general, the mechanical properties of the composites produced are substantially lower than those made of virgin carbon fibres in unidirectional arrangement. But, some differences compared to virgin fibre-reinforced plastics have to be considered:

- the nonwovens used here as reinforcement are NOT unidirectional (cf. data presented in section 3.4),
- the pyrolysed carbon fibres are without any sizing, which influences the fibre-matrix interaction, and
- rCF may be damaged by pyrolysis or mechanical processing, indicated e.g. by cracks.

Reference values are only available for virgin fibre composites. A hand lay-up and press-moulded composite based on woven fabric (i.e. bi-axial fibre arrangement, T300,Toray) with fibre volume fraction of approx. 60% exhibits tensile strength 800 MPa and Young's modulus 60 GPa 27. Almost 40% of the mechanical properties were achieved by the rCFRP reported in this work. Fibre volume fractions for nonwoven-reinforced composites are substantially lower than those 60%. From previous research it is known, that for needle-punched rCF mats the possible maximum fibre volume fraction for completely impregnated composites is not more than 50% 14. Considering the fibre volume fraction of approx. 36% for the composites reported here (i.e. 40% less than the Toray composites) the rCFRP reach a lower, but comparable level in terms of tensile strength and Young's modulus. As to be seen from the overview images in Figures 5(a), 5(c) & 5(f) for both samples containing pyrolysed fibres and exemplarily the 70 mm scraps sample, all display a good compactation with only small voids. This is also valid for the 50 mm and 100 mm scrap samples (thus not shown here).

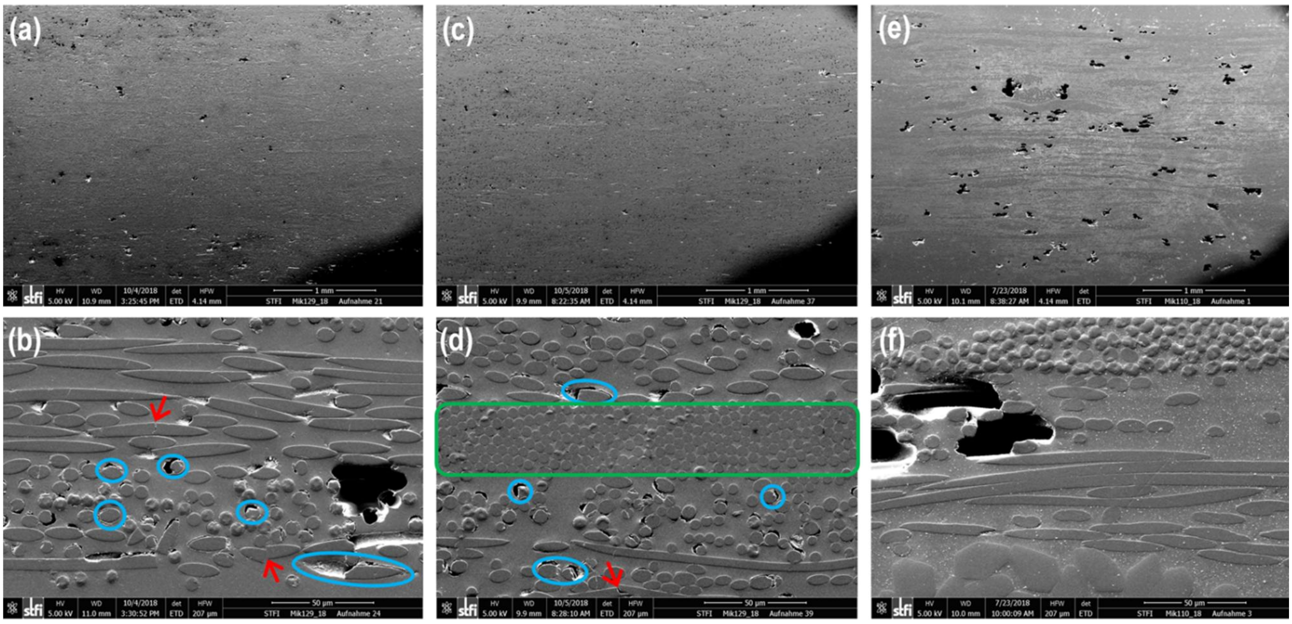

Figure 5 SEM images of the composites produced from pyrolysed rCF, (a) overview / (b) detail, 50% pyrolysed fibre/50% scraps, (c) overview / (d) detail, and scraps "70 mm", (e) overview / (f) detail. Exemplarily highlighted in (b) and (d): fibres without contact to the matrix by blue circles, cracks by red arrows, and remaining roving regions by green circles

The main difference between pyrolysed and scrap samples can be seen in the detail image Figure 5(b), highlighted exemplarily by blue circles: obviously most of the pyrolysed fibres are not impregnated well, although they are surrounded closely by the matrix. In addition, in some fibres cracks are visible, highlighted by red arrows. This coincides with the observed brittleness of the material during processing. As to expect, these effects are observed for much less fibres in the mix of 50% pyrolysed and 50% scrap fibres in Figure 5(d). Instead, this material contains larger regions of rovings not disintegrated during carding, highlighted exemplarily by green circles. These observation leads to the conclusion, that the missing sizing on the pyrolysed fibres causes a strong reduction of fibre-matrix interaction, leading to the lower mechanical strength observed for the 100% pyrolysed fibre sample.

The remaining difference of the scrap fibre samples compared to virgin fibre composites can be explained by the fibre fraction in z-direction and by the use of not endless fibres. In this context it has to be mentioned as well, that the composite production is not yet fully optimized. Thus, mechanical properties of approx. 80% of virgin fibre composites seem to be achievable for identical fibre volume fractions.

3.2 Composites with different fibre volume fractions using RTM

Composites with different fibre volume fractions have been produced on site of project partner SCHMUHL using the resin transfer moulding (RTM) process. Production scraps of SCHMUHL have been processed at STFI into nonwovens of 300 g/m², using the carding/cross-lapping process with subsequent needle-punching. Composite boards of 700 x 700 mm² with fibre volume fractions from 11% to 27% have been produced from this rCF-nonwoven using the RTM process. Epoxy resin was used as matrix.

Figure 6 displays the tensile strength of the composites. It can be clearly observed that again the tensile strength in CD is higher than in MD, due to the process variant with carding and subsequent cross-lapper. As to expect, the tensile strength increases with increasing fibre content in the material. As for the scrap samples described in the previous section, only a negligible number of small voids was detected in these composites by SEM, giving no hint for differences in impregnation.

The corresponding values for Young's modulus are presented in Figure 7, displaying the same tendency with Young's modulus dependent on increasing fibre fraction. Flexural strength and modulus of these samples follow the same scheme and are thus not shown here.

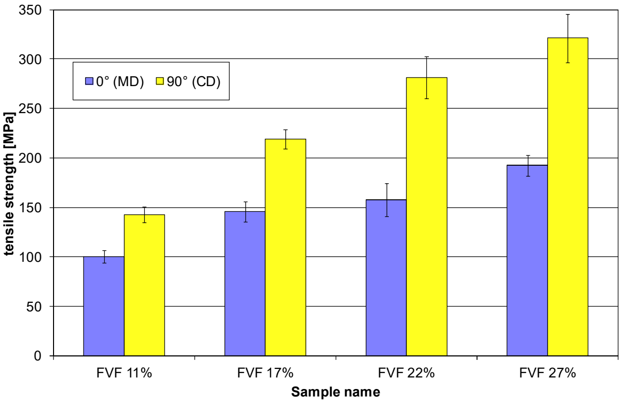

Figure 6 Tensile strength of the composites produced from rCF with different fibre volume fractions

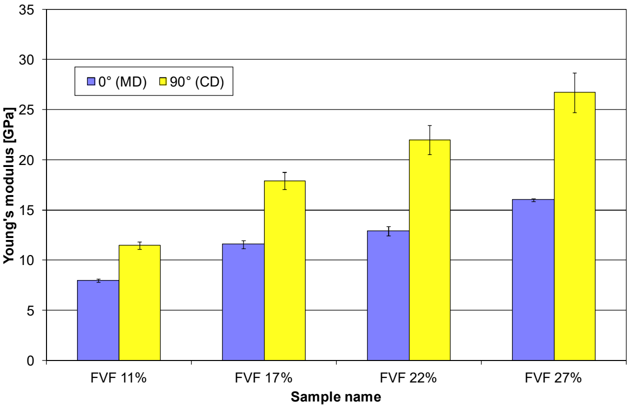

Figure 7 Young's modulus of the composites with different fibre volume fractions

3.3 Composites made of rCF-prepregs

Two rCF-prepregs have been produced at c-m-p GmbH, Heinsberg, DE from rCF-nonwovens (i) delivered by TENOWO (5731/3, Zetacomp CF, 266 g/m², based on scrap fibres) and (ii) STFI (K12, 300 g/m², based on scrap fibres). The nonwoven processed at TENOWO was carded and stitch-bonded, whereas the STFI-nonwoven was processed by airlay and needling. The applied matrix content was 67% epoxy resin 'CP041'. Both prepregs were consolidated in two variants: (i) press-moulding at STFI and (ii) autoclave method at EISSMANN COTESA. As reference, both nonwovens were laminated in hand lay-up at STFI and press-moulded. The samples and processing variants are listed in Table 3.

Table 3 Prepreg samples and processing variants

Sample name	Specifications	
	Process	**FVF [%]**
5731/3 HL	hand lay-up, press-moulding (reference sample)	38.2
5731/3 V1	prepreg, press-moulding	41.4
5731/3 V2	prepreg, autoclave method	17.0
K12 HL	hand lay-up, press-moulding (reference sample)	33.6
K12 V1	prepreg, press-moulding	37.0
K12 V2	prepreg, autoclave method	20.5

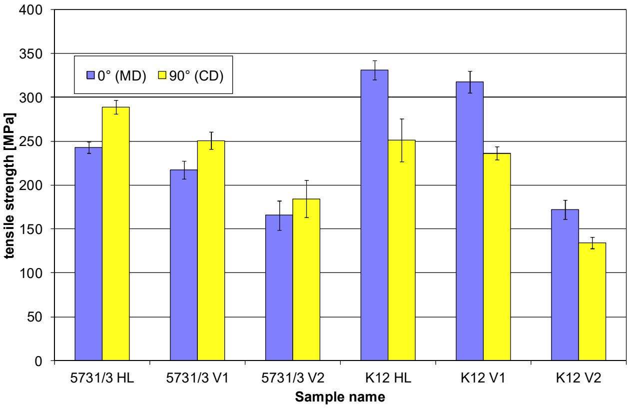

Figure 8 Tensile strength of the composites produced from rCF prepregs

The tensile strength determined for these composite boards is displayed in Figure 8.

Composites based on carded nonwovens show a significant CD orientation due to the carding and cross-lapping process, in contrast, the composites based on airlay nonwovens show a higher MD orientation. According to modelling of the airlay process 28 the higher MD orientation can be attributed to a higher conveyor belt speed.

For both composite variants (carded and airlay based) the tensile strength of the hand laminates is higher than for the prepreg samples. SEM analysis revealed that the hand laminates display the same amount of voids like the press-moulded V1-samples. Thus, the slightly lower tenacity of the press-moulded variants is caused more by the different resin used for V1 and V2.

The different lamination processes have also be condsidered: in hand lay-up each nonwoven layer in the stack is impregnated individually, while in prepreg production the resin is applied from one side. This may cause a worse impregnation of the nonwoven and, thus, a lower tensile strength of the composites. Presumably, the worse impregnation of the nonwoven can be compensated better by press-moulding, where mechanical pressure forces the resin to impregnate the nonwoven. Consequently, both V1-samples show a tensile strength similar to the reference, whereas the V2-samples made by autoclave process display a significantly lower tensile strength. Especially the airlay based composites show a strong decrease (approx. 46%), while carding based composites show a loss of approx. 27%. This could be partly due to the technique of the airlay process with less fibre opening.

Not fully opened, unseparated fibres lead to higher restoring forces (due to the missing separation and orientation) and a larger number of air pockets that impede the impregnation process. While the impregnation using prepreg technology combined with a hydraulic column down-stroke press enables high pressure and slow heating up with constantly additional pressure, the prepreg technology combined with autoclave draws vacuum and cures the matrix material. This difference of the two technologies and the one sided impregnation causes probably more pipes and less impregnation during the autoclave process due to the resin flowing properties. The obtained SEM images are displayed in Figure 9 and provide this assumption.

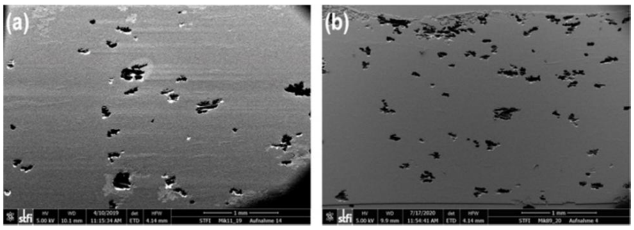

Figure 9 SEM images of composites based on airlay nonwovens produced using (a) press moulding and (b) autoclave technology

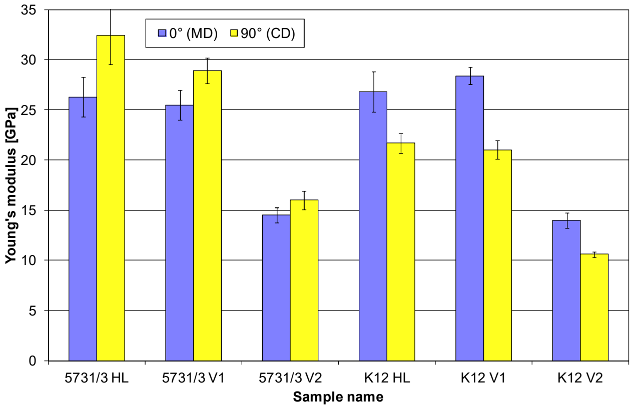

Figure 10 Young's modulus of the composites produced from rCF prepregs

In Figure 10 the corresponding Young's modulus values are shown. They display the same tendencies. This is also valid for the flexural strength and -modulus of these samples (not shown here). In general, these results display the feasibility of the prepreg process for rCF nonwovens.

Due to the achieved fibre volume fractions of approx. 10-38% (depending on the impregnation method, c.f. Tables 2 & 3) several demonstrators were developed (described in 29). At EISSMANN Cotesa, for example, a spoiler based on prepregs of stitch-bonded rCF-nonwovens combined with woven fabrics was developed. The spoiler is consolidated in autoclave method.

3.4 Composites from re-recycled carbon fibres

A lot of scraps (roving snippets, length 120 mm) has been used to test the ability of the process for repeated recycling. Nonwovens have been produced from this lot using both process variants (carding and airlay) with subsequent needle-punching.

Table 4 Composites produced from rCF and rrCF nonwovens

Sample name	Specifications			
	nonwoven grammage [g/m²]	MD/CD ratio by on-line monitoring	no. of layers in composite	FVF [%]
rCF-Airlay	300	0.92	6	37.0
rrCF-Airlay	250	n/a	7	26.3
rCF-carded	300	0.30	6	34.6
rrCF-carded	100	0.88	18	28.9

A part of this rCF nonwoven has been supplied again to tearing and nonwovens production as described above. The fibre length was reduced from initially 119 mm (incoming control) via 104 mm (first tearing) to finally 69 mm (second tearing). In the second process it was not possible to reach the intended grammage of 300 g/m². Due to the reduced fibre length, the resulting weight of the nonwovens was 250 g/m² in airlay and 100 g/m² in carding process. The resulting, so-called 'rrCF nonwovens', and the corresponding rCF nonwovens have been processed as usual by impregnation with epoxy resin and press-moulding. The samples and processing variants are listed in Table 4. An increasing number of layers in the composite lead to higher volume between the layers, which increases the thickness of the composite and decreases the FVF (Table 4).

Figure 11 shows the tensile strength of the different samples. For both airlay variants the results are as expected: tensile strength approx. 300 MPa and close to isotropic distribution. The MD/CD ratio calculated from the tensile strength is for both of them 0.8. The corresponding value from on-line monitoring at the line for rCF-Airlay is 0.9. Due to the reduced fibre length, the tensile strength of the re-recycled variant (rrCF-Airlay) is slightly lower.

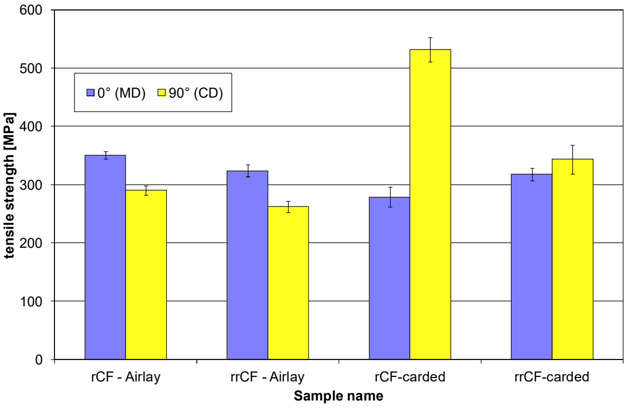

Figure 11 Tensile strength of the composites produced from rCF and rrCF nonwovens

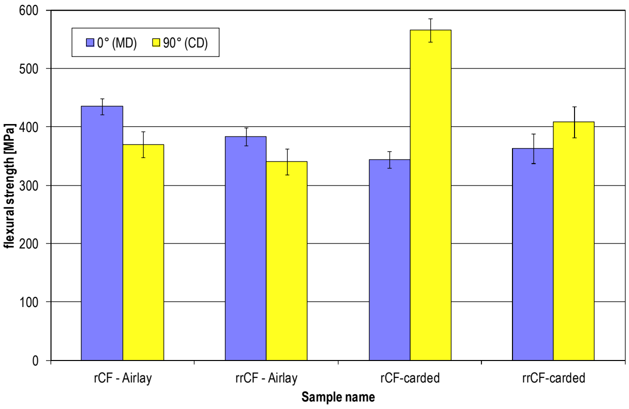

Figure 12 Flexural strength of the composites produced from rCF and rrCF nonwovens

For the carded samples the situation is different: the carded rCF sample displays the expected anisotropy with predominant tenacity in CD. The values of 279 MPa for MD and 532 MPa for CD conform to a MD/CD ratio of 0.5, whereas the value of on-line monitoring is 0.3. The re-recycled variant rrCF-carded displays unexpectedly a quasi-isotropic behaviour: the tensile strength is 318 MPa for MD and 343 MPa for CD, corresponding to a MD/CD ratio of 0.92. The value of on-line monitoring (0.88) supports this finding.

The flexural strength of these samples is presented in Figure 12, displaying exactly the same tendencies. This is also valid for Young's modulus and flexural modulus of these samples (not shown here).

In addition, the SEM image of the carded rrCF sample (Figure 13) does not exhibit unexpected findings. As for the other hand-lay-up samples, there are few voids to observe, while the impregnation is good. The only difference is that the rrCF display some cracks, as highlighted by red arrows. These cracks must be caused by the carding process prior to impregnation, because they are completely filled with matrix material.

The original data from on-line orientation analysis (previously published in 19) are presented in Figure 14 as histogram for the carded rCF and rrCF samples. It is easy to observe, that the carded rCF sample shows a clear CD-orientation with maximal fibre shares oriented around 90° and –90°. In opposite to that the rrCF sample is nearly homogeneously distributed, indicating a quasi-isotropic fibre orientation.

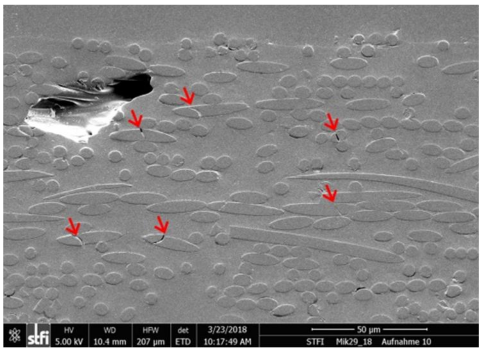

Figure 13 SEM image of the carded rrCF sample

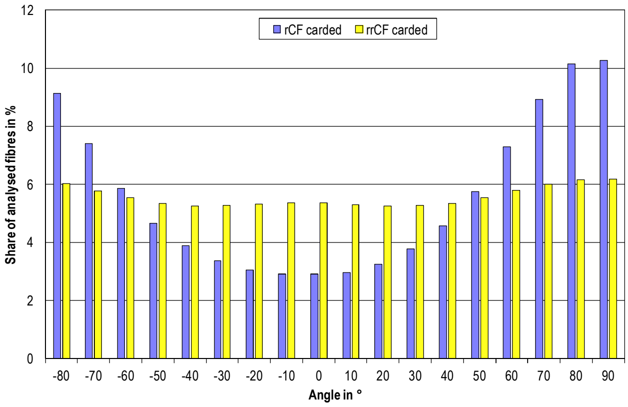

Figure 14 Distribution of fibre orientation from on-line analysis after carding process for rCF and rrCF. Data from 19, modified presentation

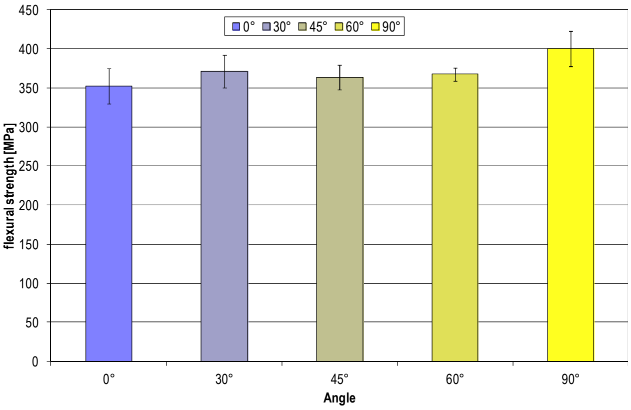

Figure 15 Flexural strength of sample rrCF-carded depending on the angle

In order to validate the nearly isotropic structure of this sample, additional specimens were prepared to analyse the tenacity in 45°. The result was 304 MPa, i.e. in the same range like the 0° and 90° samples (317 & 343 MPa). Due to the limited board size it was only possible to prepare smaller specimen for flexural strength analysis in 30°, 45° and 60° to analyse the properties more in detail. The results are shown in combination with the already known values for 0° and 90° in Figure 15. Visually there is a small but not significant increase from 0° to 90°, but in general the values are within the range of uncertainty. Thus, the material is really quasi-isotropic, as also seen from the on-line-control (cf. Table 3 & Figure 14).

This effect is caused by the higher haul-off speed in the cross-lapper, leading finally to a fibre orientation close to that from the airlay process 19.

In general, the results show the feasibility of at least one additional recycling cycle for carbon fibres with only minor loss of composite stability.

4 CONCLUSION

A process line has been set-up successfully, enabling the processing of 100% recycled carbon fibres into high-quality nonwovens by carding as well as by airlay. As reported elsewhere, in parallel a quality control system has been defined to ensure constant product qualities 19. In combination, this offers the possibility to produce taylor-made rCF qualities. Various lots of rCF, ranging from production scraps to pyrolysed fibres from 'end-of-life' parts, have been processed successfully to carded and airlay nonwovens. The possible range of fibre orientation after carding process reaches from highly-oriented in CD to nearly isotropic, depending on the material and processing parameters.

rCF-reinforced plastics have been produced, using different typical industrial processes to prove the processability. In press-moulding tensile strength values of approx. 530 MPa in CD and 330 MPa in MD have been achieved, combined with Young's moduli of 50 GPa (CD) and 31 GPa (MD). Similar values were obtained by resin-transfer moulding (RTM).

Using stitch-bonded nonwovens, the production of prepregs opened successfully another process pathway. The prepreg-based composites displayed tensile strength up to 330 MPa and Young's modulus up to 29 GPa.

In addition, first experiments showed that repeated recycling of carbon fibres is possible. The composites produced of so-called 'rrCF' displayed only a minor loss of stability compared to the corresponding rCFRP.

Compared to composites based on woven virgin-fibre fabric with fibre volume fraction of approx. 60% 27, almost 40% of the mechanical properties were achieved by the rCFRP reported in this work. Considering the lower fibre volume fraction of approx. 36% the rCFRP reach a lower, but comparable level in terms of tensile strength and Young’s modulus. This difference can be explained not only by the fibre fraction in z-direction and by the use of not endless fibres. As well it has to be mentioned, that the rCF composite production is not yet fully optimized. Thus, mechanical properties of approx. 80 % of virgin fibre composites seem to be achievable.

Another topic for discussion is the price of rCF compared to virgin fibres: it is lower, ranging from 5 €/kg for scraps up to 12 €/kg for pyrolysed fibres in 2017. The future forecast for pyrolysed fibres was a decrease to 7 – 9 €/kg with increasing production capacities 14.

Summed up, the carbon fibre nonwovens generated on the line are very suitable as semi-finished products for CFRP-structures. Due to their high formability sufficient strength, and adjustable MD/CD ratio they are ideal candidates for sophisticated composites. Summed up, an industrial feasible process for recycling of carbon fibres has been proven to operate.

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GEFÖRDERT VOM

zwanzig20
PARTNERSCHAFT FÜR INNOVATION

Bundesministerium
für Bildung
und Forschung

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