RECYCLED CARBON-FIBER FLEECES AS A GREEN ALTERNATIVE FOR REINFORCING IN-PLANE MECHANICAL PROPERTIES OF THERMOSET COMPOSITES

A. Rios de Anda[a*], T. Neumeyer[a], H. Lengsfeld, T. Rademacker[b], V. Altstädt[a]

[a]*Department of Polymer Engineering, University of Bayreuth, Universitätsstr. 30, 95447 Bayreuth, Germany*
[b]*CarboNXT GmbH, Stader Str. 55-63, 21737 Wischhafen, Germany*
** agustin.rios-de-anda@uni-bayreuth.de*

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Abstract

The reinforcement capabilities of CarboNXT[1] 100% individuel recycled carbon fibers were considered in this research. Two types of fleeces were herein studied: Stitched-bonded and Non-Stitched veils. Laminates were obtained by RTM with a rapid-curing two-component epoxy resin as a matrix. The recycled carbon fibers exhibited a good processability. Mechanical in-plane properties were investigated. It was observed that for an equivalent fiber volume content, stitched-fiber composites exhibited higher moduli and lower strengths than the non-stitched ones. We believe that this might be due to the presence of out-of-plane oriented fibers in the non-stitched fleece.

[1] *supplied by* CarboNXT GmbH

1. Introduction

A novel carbon fiber recycling process has been recently developed. Little is known on how this process affects the obtained recycled carbon fibers on their processing characteristics and their reinforcement capabilities in thermoset composites. CarboNXT GmbH specializes in obtaining recycled fiber veils from polymeric composite parts that have reached their life cycle. This company has developed an oxidizing pyrolysis process in which the out of usage composite parts are treated to obtain long fibers, which are then woven into veils. In this work, the relationship between processability, material inner structure, and mechanical properties of composite materials reinforced with such recycled fibers was investigated. A rapid-curing two-component resin used widely in the automotive industry was considered as a matrix. Firstly the processability of two different veils, Stitched-bonded and Non-Stitched, was studied. Composite material laminates were obtained by resin transfer molding (RTM). It was observed that both veils are well-processable and fiber volume contents of up to 26 % in the composite can be easily obtained. Afterwards structure property relationships between the fiber volume content and fleece style with the mechanical properties of such laminates was established. The characterized in-plane mechanical properties are the tensile, compression, and bending properties, which are relevant in industrial applications. We attempt to understand in this paper the reinforcement capabilities of such fibers and the in-plane mechanical properties through their nature and architecture.

2. Experimental

2.1. Materials and Processing

CarboNXT *100 % individuel* recycled carbon fibers provided by CarboNXT GmbH were considered in this research. Two types of fleeces were studied: stitched-bonded and non-stitched-bonded veils. Both fleeces have an aerial weigh of 200 g/m^2. Figure 1 shows the fiber distribution and orientation of these veils as observed by optical microscopy.

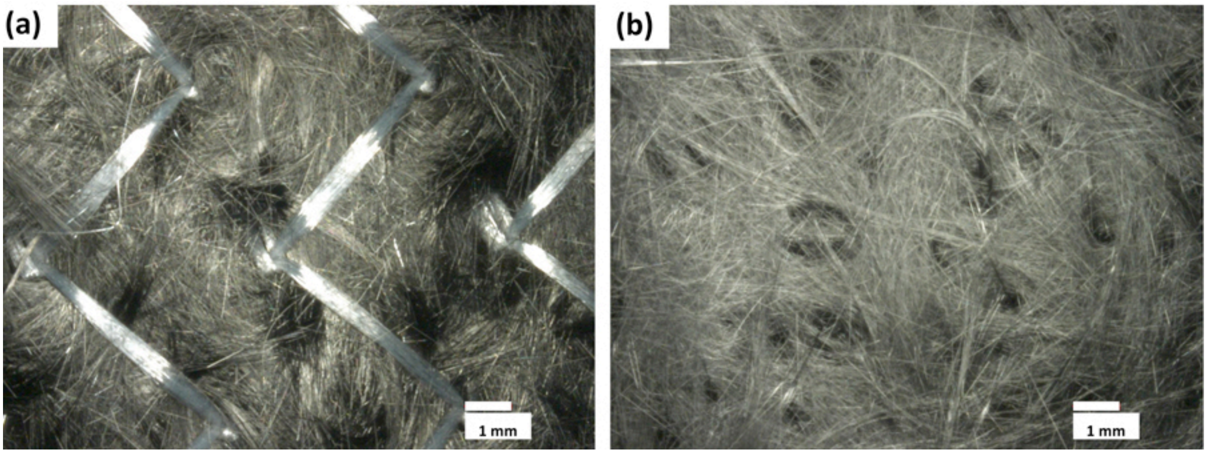

Figure 1. Optical microscopy pictures (magnification × 25) showing the fiber distribution and orientation in the (a) stitched-bonded and the (b) non-stitched CarboNXT recycled carbon fiber fleeces.

In order to produce 2 mm-thick (theoretical) laminates, six plies of each of these fleeces were stacked up in the mold before injecting the resin. The number of layers yield the maximum possible fiber volume content (*FVC*) as the fleeces cannot be further compressed. For this thickness the theoretical *FVC* is of 26 $\%_{vol}$ *was reached*. Two laminates were produced by RTM using a Momentive® EPIKOTE™ Resin 05475 / EPIKURE™ Curing Agent 05443 two-component system. This system is widely used in the automotive industry. The resin and the hardener were kept at 60 ºC and degased for 10 minutes prior to injection. The mixture weight ratio was set at 100 : 24 resin/hardener. Both the resin and the hardener were injected with a set pressure of 5 bars at 90 ºC. These two components were combined in a static mixture just before the line entering the RTM mold. The resin / hardener injection time was about 2 minutes for both laminates. After the injection was finished, the mold was heated up to 120 ºC with a set temperature ramp of 4 ºC/min, and then the resin was cured at this temperature for 10 minutes. After curing, the laminate was cooled down to room temperature with a set temperature ramp of 4 ºC/min as well.

2.2. Samples and Testing

Ultrasonic characterization was carried out with a Dr. Hillger HFUS2400 apparatus to determine the processability of the fleeces. C-Scan thickness (volume) and back-surface signals were taken with a frequency of 5 MHz. No impurities, voids, or defects were observed in the laminates. Dynamic Mechanical Analyses were conducted to measure the glass transition temperature (T_g) of the laminates. Two samples per laminate of dimensions $50 \times 10 \times 2$ mm^3 were measured with a TA Instruments RDA III in torsion mode. Samples were heated from 25 to 200 ºC with a 2º C/min ramp and tested with a frequency of 1 Hz and a deformation amplitude of 0.1%. The values of T_g measured for both laminates, taken at the maximum of *tan δ*, are of 127±2 ºC for the stitched laminate and of 129 ± 2 ºC for the non-stitched one. These values correspond well to that stated in the Momentive resin datasheet (T_g = 130 ºC) [1].

Mechanical tests at room temperature were conducted to obtain the tensile, bending, and compressive properties of the laminates. Six samples per test per laminate were considered. Three-point bending tests were carried out for the three series of samples with a Zwick Roell Z2.5 machine equipped with a force sensor of 2.5 kN. DIN EN ISO 14125 standard guidelines were used to test such samples. Samples of 100×15×2 mm^3 were tested. The lower beam distance was set at 80 mm and the samples were tested with a load rate of 1 mm/min. Tensile tests were conducted with a Zwick Roell Z1475 machine equipped with a force sensor of 100 kN. DIN EN ISO 527-4 was considered for testing the samples. Samples of 250 × 25 × 2 mm^3 were considered. Glass-fiber reinforced epoxy resin end-tabs of 25 × 50 mm^2 were glued to the extremes of each sample from both sides. Strain gages (HMB Type: 6/120 LY13) were glued in the middle of the sample, one on each side, to precisely record the elongation during the measurements. Samples were tested with a strain rate of 2 mm/min. Finally, Celanese compression tests were also done with the Zwick Roell Z1475 machine. Samples of 112 × 10 × 2 mm^3 were tested according to the DIN EN 2850 standard. End tabs of 10 × 51 mm^2 were glued as with the tensile specimens. Also, strain gages were glued at each side (HMB Type: 3/120 LY13) as well so as to measure the elongation during the tests. Samples were compressed with a rate of 1 mm/min.

3. Results and Discussion

3.1. In-plane elastic modulus

The elastic moduli obtained experimentally were compared to theoretical values given by the Manera and the Pan models [2-4] for 2- or 3-dimension isotropic-distributed fibers. We consider a 2-dimension isotropic distribution for the theoretical calculations of the fibers. This is achieved due to the process for obtaining fiber mats.

The Manera model [2,3] considers a geometric random isotropic fiber distribution which does not depend on the fiber volume content. This model can only be considered for fiber volume contents ranging from 10 to 40 $\%_{vol}$ and for neat resins having an elastic modulus between 2 and 4 GPa. Both our fiber volume fraction and resin modulus comply with these criteria. The resulting modulus is calculated as shown in Equation 1:

$$E_C^{2D} = \left(\frac{16}{45}E_f + 2E_m\right) + \frac{8}{9}E_m \tag{1}$$

The Pan model [2,4] takes into account a radial isotropic distribution that depends on the fiber volume content. This model can be used without restrictions regarding the fiber volume content or the neat matrix elastic modulus. The resulting composite modulus can be calculated according to Equation 2:

$$E_C^{2D} = \frac{V_f}{\pi}E_f + \left(1 - \frac{V_f}{\pi}\right)E_m \tag{2}$$

For both models E_C^{2D}, E_f, and E_m correspond to the modulus of the composite, the fibers, and the matrix respectively; whereas V_f is the fiber volume content in the composite. The value of E_m was taken from the Momentive system datasheet [1].

Furthermore, in order to compare the elastic moduli obtained experimentally, the moduli values were normalized to a *FVC* of 26 %. This is done since the laminates are not exactly 2 mm thick (i.e. 2.2 mm) and the actual *FVC* is lower. Figure 2 shows the tensile, compression, and bending moduli normalized to a FVC of 26 % for the stitched and non-stitched laminates. The plotted values are compared to the moduli calculated from the Manera and Pan models.

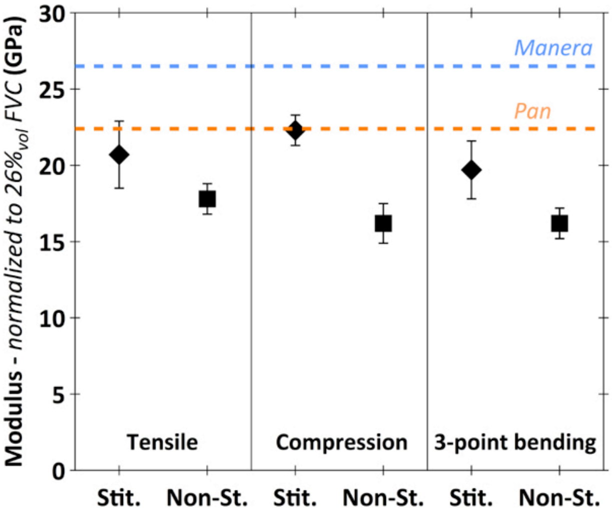

Figure 2. Tensile, compression, and three-point bending elastic moduli normalized to a *FVC* of 26 % for both stitched and non-stitched laminates. Values are compared to the moduli calculated from the Manera and Pan models [2-4].

We firstly observe in Figure 2 that the experimental elastic moduli, especially for the stitched fleeces, are of the same order of magnitude as the ones predicted by the Pan model. This would mean that these fibers are effectively capable of yielding a good reinforcement within a resin matrix, as predicted for a 2-D long fiber veil. Moreover, it is seen that the stitched fleeces exhibit higher moduli values than the non-stitched ones. We might think of two hypotheses to explain this effect. One hypothesis could be, that the difference in these values could come from an influence of the fiber orientation in the fleeces. It could be possible that in the case of non-stitched fleeces some fibers would be oriented in an out-of-plane direction, which would not be present in the stitched ones as the stitching would orient them in the in-plane direction. The other hypothesis comes from the fact that the non-stitched fleeces are pinned. The pinning produces small holes (of ca. 1 cm in diameter) in the fleece, as shown in Figure 1. It might be possible that these holes are still present in the injected laminate, and that are matrix-rich regions with few fibers. If this were so, these matrix-rich regions would act as stress concentrators and would degrade the overall samples properties, as their local mechanical resistance would be lower. These hypotheses would explain why the elastic moduli of the non-stitched laminate are smaller, although they have to be yet confirmed.

3.2. In-plane ultimate strength

Figure 3 shows the normalized strength values obtained by tensile, compression, and three-point bending tests carried out on the stitched and non-stitched laminates. We notice that the strength values of the Non-Stitched samples are higher than those of the stitched ones. This is somehow surprising as the in-plane mechanical properties of the Non-Stitched laminate are lower than those of the Stitched samples. As with the elastic moduli, we could only think that this phenomenon might be explained by either a variation of the fibers architecture and/or orientation or the presence of matrix-rich domains within the laminate in the case of the Non-Stitched veils.

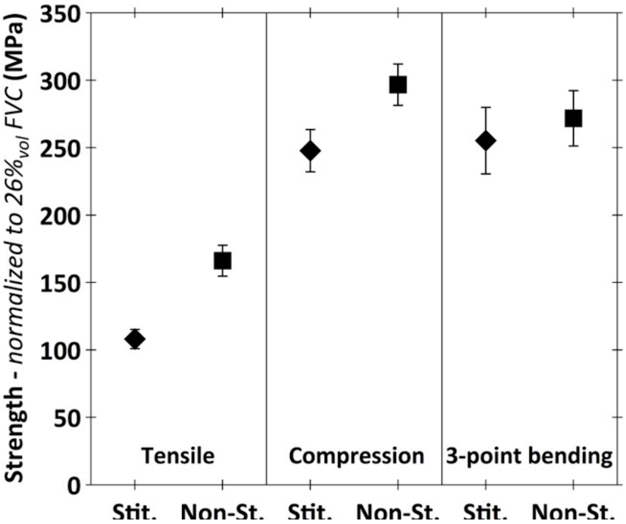

Figure 3. Tensile, compression, and three-point bending normalized strength measured for both stitched and non-stitched laminates.

3. Conclusions and Perspectives

The processability and reinforcement capabilities of novel recycled carbon fibers were studied in this work. Two types of veils, stitched-bonded and non-stitched, were considered. Two laminates, each reinforced with the mentioned fleeces and aimed for in-plane mechanical testing, were hereby obtained by RTM using a two-component matrix system. Ultrasound and DMA quality characterizations showed that the veils are well-processable by RTM, yielding homogeneous laminates. Afterwards the tensile, compression, and bending in-plane properties were characterized by mechanical tests. Concerning the elastic moduli, it was observed that the composites, especially the Stitched-fiber one, had close values to those predicted to numerical models. Moreover the non-stitched-fiber composites exhibited lower values than those of the stitched-fiber ones. Concerning the ultimate strength the non-stitched-fiber composites exhibited larger values, which is quite surprising since their elastic moduli are smaller. We believe that these two results might be due to either the non-stitched fleeces having fibers oriented in an out-of-plane fiber direction, or the presence of matrix-rich domains in the non-stitched laminates as these fleeces present pinning holes due to their obtaining processing. Further studies, notably concerning the investigation of out-of plane properties such as impact and damage resistance, as well as a microscopy characterization of the fiber length distribution and orientation in the composites will be carried out to better elucidate and understand the reinforcement properties of these recycled carbon fibers.

4. Acknowledgements

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