COMPARISON OF INTERLAMINAR AND INTERFACIAL SHEAR STRENGTH WITH RECYCLED CARBON FIBER

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Abstract: *Testing the interlaminar shear strength (ILSS) of composites using a short beam shear (SBS) test takes time. Micro-scale interfacial shear strength values (IFSS) need to be scaled back to the macro-level if a more reliable picture of the composite behavior on the right scale is desired. Here, the objective is to find out what kind of relation the ILSS and the IFSS values have for recycled carbon fibers (rCF) and epoxy matrix. Factors affecting the reliability of both tests are considered.*

A slope-based method was used to obtain the IFSS values from the micro-scale results. The microbond test gave a 43.6 MPa IFSS value and the SBS test a 36.4 MPa ILSS value. There were a lot of variables associated with the SBS for rCF, due to which there is uncertainty in the results. The data from the microbond tests were more reliable due to the high sampling volume.

Keywords: ILSS; IFSS; recycled carbon fiber; short beam shear test; microbond test

1. Introduction

The importance of composite recycling has increased, which can be noticed in the legislation of the European union considering recycling and circularity. Pyrolytic recycling has been developed to recover carbon fiber, but it is known to have a detrimental effect on the mechanical properties of the fibers [1]. For recycled carbon fiber (rCF) to meet the mechanical requirements of the reuse application, the properties of different recycling batches should be tested reliably and quickly. The established method to test interlaminar shear strength (ILSS) of composites is the short beam shear (SBS) test, but there are problems [2] considering the test, such as identifying the failure mode of the sample and thus validating the result for ILSS. In addition, making the laminate samples takes time. The different manufacturing steps increase the possibility of distortion of the fabric and therefore deviation of the results. Testing the micro-scale properties and reliable scaling of results to the macro level will be able to save hours and avoid possible errors [7].

In this study, macro and micro-scale shear strength is compared for a rCF and reinforced epoxy matrix. Macro-scale data was obtained by SBS and ILSS values were calculated. Micro-scale data and the interfacial shear strength (IFSS) were analyzed by the slope-based approach of microbond test. Questions, how the traditional ILSS values relate to IFSS values, and what magnitude the results, are answered. The reliability of the results is also considered.

2. Materials and methods

2.1 Recycled carbon fiber and matrix

The original carbon fiber roll [4] (Toho Tenax, Japan) was pre-impregnated with epoxy, where the weight percentage of carbon fiber was 70%. Recycling was done using pyrolysis as part of the previous study conducted by Palola et al. [4]. The yield was 67% and the virgin carbon fiber fabric's weave was Twill. Other characteristics of the carbon fiber are noted in Palola's et al. publication [4].

The matrix was epoxy Araldite® LY 5052 and hardener Aradur® 5052 CH (Huntsmann, USA) and they were mixed by weight in a ratio of 100 to 38, respectively, as recommended by the manufacturer [5].

2.2 Short beam shear test

SBS samples were manufactured using vacuum assisted resin transfer infusion (VARI). Eight plies of rCF were dried in oven for 30 min at 100 °C and stacked in sequence of [0°$_8$]. Vacuum pressure of 0.6 bar was used. The laminate was cured at room temperature for 20 h and post-cured for 12 h at 60 °C. The ILSS was tested using a universal testing machine (Instron 5967, UK) according to standard ASTM D 2344 [7]. Nine samples were cut to approximate dimension of 12.5 mm x 4.2 mm with circular saw (Proxxon, Germany), which had a diamond coated blade. Average thickness for the samples was 2.14 mm. Three-point bending configuration was used with span length of 5.67 and displacement rate of 1.0 mm/min. The samples were pre-tensioned to 3 N to get the same initial set-up. ILSS is calculated using the Eq. (1):

$$ILSS = \frac{3}{4}\frac{F_{max}}{b*h}, \quad (1)$$

where F_{max} is the maximum force in the three-point bending, b and h are the sample thickness and width, respectively. The fiber weight percentage was calculated to be 59.4 % for the laminate.

2.3 Microbond test

The FIBRObond micro testing device (Fibrobotics, Finland) was used for microlevel adhesion testing [7] [8] to get IFSS. The operator disconnects individual epoxy droplets from the fiber by controlling the blades with piezoelectric locators. Figure 1. shows an illustrative picture of fiber, droplet, the sample holder, as well as the blades with which the droplet is removed.

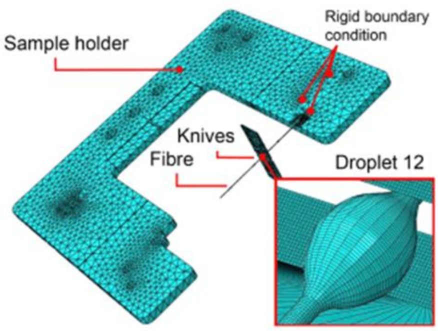

Figure 1. FIBRObond testing configuration [9].

The fibers were selected from a carbon fiber fabric area that represented the entire fabric. The droplets were prepared for six fibers after which they were cured at ambient laboratory temperature for 60 h. The same epoxy was used for laminates. The force-displacement curve of

  doi:10.5075/epfl-298799_978-2-9701614-0-0

each droplet was recorded with a 1 N force cell (Futek jr. S-beam, USA) and droplet length was measured using machine vision. The displacement rate was 0.008 mm/s. The six individual rCF filaments were tested and each filament contained 8 - 40 tested droplets. To calculate IFSS, a slope-based approach was used, where the maximum force to detach each droplet is drawn on the graph as a function of the droplet's surface area. Linear regression fit is then used to get IFSS. The microbond data is further discussed in [4].

2.4 Scanning electron microscopy

To ensure the fracture mode of the SBS samples were interlaminar, a scanning electron microscopy (SEM, Zeiss ULTRAplus, Germany) was used. Pictures of the fracture surfaces were taken from the samples L6, L7 and L8 (see Table 1.). In addition, one cross-sectional sample was prepared from the laminate to ensure the matrix's adhesion to fibers is sufficient. The cross-section sample was cut from the laminate using a diamond coated blade and was cast in transparent resin EpoFix (Struers, Denmark) in vacuum impregnation unit CitoVac (Struers, Denmark). A vacuum pressure of 0.2 bar was used. After curing at room temperature for 4 days the cross-section sample was sanded with water using #500, #1000, #2000, and # 4000 grit paper and polished with polishing cloth and 3 µm and 1 µm diamond suspension in succession. The SEM samples were coated with a ~3 nm layer of palladium and platinum mixture (ratio: Pt/Pd 80/20 wt%).

3. Results and discussion

3.1 Interlaminar shear strength

The SEM images confirmed the matrix had wet the fiber and the laminates were of high quality (see Figure 3. a) and 3.b)). The dominant fracture mode for the tested samples was noted to be an interlaminar fracture in the three samples imaged.

In Figure 2. are the SBS samples' flexural stress and strain behavior. The samples L4 and L9 had 30 % force drop during the test, which caused the test to stop. This limit was user-defined according to standard ASTM D 2344 [7]. Samples L5 and L8 have a different flexural behavior after 1 mm of flexural elongation compared to other samples as seen in Figure 2. This can be explained by the difference in the thicknesses. L5 is thicker (2.31 mm) and L8 is thinnest (2.05 mm).

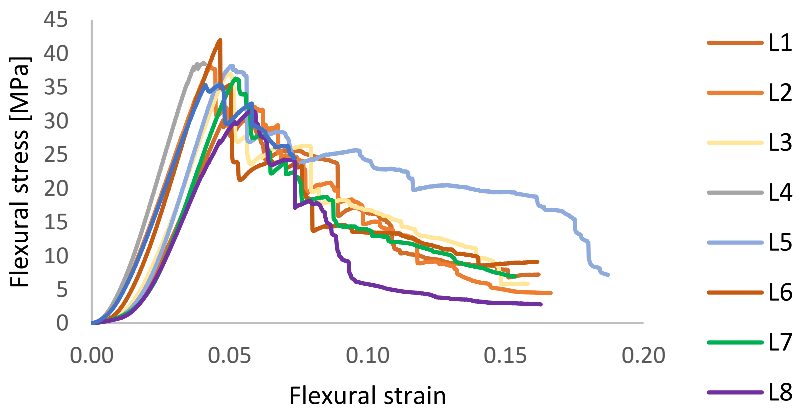

Figure 2. ILSS samples' flexural stress as a function of flexural strain.

Table 1: SBS samples' dimensions, ILSS values, mean (x), standard deviation (s) and coefficient of variation (CV).

Sample	Lenght [mm]	Thickness [mm]	Width [mm]	ILSS [MPa]	x [MPa]	s	CV [%]
L1	12.59	2.15	4.37	30.28			
L2	12.67	2.13	4.35	38.07			
L3	12.57	2.13	4.35	37.03			
L4	12.45	2.08	4.10	38.59			
L5	12.50	2.31	4.07	38.18	36.36	3.62	9.96
L6	12.46	2.14	4.20	42.00			
L7	12.35	2.03	4.00	36.28			
L8	12.48	2.05	4.09	31.53			
L9	12.81	2.24	4.24	35.32			

The laminate had abnormal epoxy-free area that is depicted in Figure 3. c). The lowest carbon fiber layer was distorted during layup or infusion. Whether there are distortions in other layers of the laminate was unknown. However, this is one good example of the challenging sample preparation of the SBS test with rCF. The uneven quality and the surface of the fiber can cause uneven distribution of the load in the sample and increase the likelihood of stress concentrations [10]. In this case, the laminate is also more likely to break under the load when compared to virgin carbon fiber. Palola et al. observed surface unevenness on individual rCF [4].

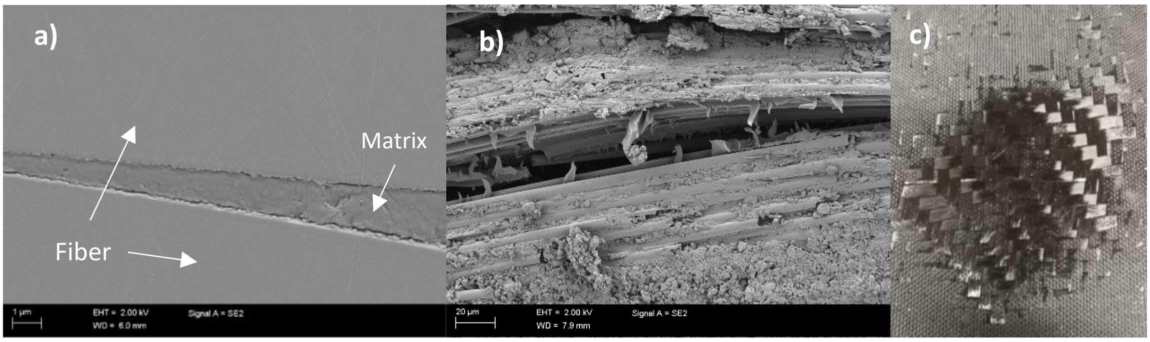

Figure 3. a) SEM image from the cross section of the laminate, b) fracture surface with interlaminar debonding, and c) distorted ply where there is little to no resin.

Factors affecting ILSS are, among other things, carbon fiber fabric weaving, reinforcement layup, and thickness [2]. Carbon fibers have low surface activity and adhesion to resins, which decreases ILSS [10] and IFSS. Also, the recycled carbon fiber fabric was not of the best quality. Instead, there were obvious gaps, which are due to the method of recycling the carbon fiber. Since carbon fiber fabric weaving was not the best possible the poor quality might have had some effect on ILSS, although good areas of fabric were selected visually for the laminate plies.

3.2 Interfacial shear strength

The IFSS was 43.6 MPa and had a standard deviation of 2.5 MPa [4] (see Table 2.). Palola et al. conducted the microbond tests on rCF and analyzed the results and considered the factors that contributed to them in their study. The strength of recycled carbon fiber had been eroded in recycling because the embedded length of the largest droplets the fiber withstood during the test was 42 µm. Palola et al. noticed a 60 % drop in tensile strength of the rCF compared to virgin carbon fiber [4]. The FIBRObond measuring device was able to achieve good linear regression, which the disturbance term (R^2) indicates, although the rCF had significantly lower tensile strength.

Laurikainen et al. [8] address the high scatter and error sources of microbond testing in their study, as well as the factors contributing to these. The high scatter of the microbond test was largely due to variation in the quality of fibers and the quality of the fiber surfaces, rather than the FIBRObond measurement apparatus itself. The total number of droplets they tested was 1527.

Table 2: The results of the microbond test for rCF with number of tested droplets, IFSS values, disturbance terms (R^2), mean (x) and standard deviation (s) [4].

Sample	Number of tested droplets	IFSS [MPa]	R^2	x [MPa]	s
M1	8	41.5	0.89	43.6	2.5
M2	11	40.6	0.93		
M3	44	37.6	0.95		
M4	31	50.9	0.97		
M5	43	41.2	0.95		
M6	40	49.7	0.98		

3.3 Comparison between micro and macro-scale

Measured ILSS was 16.6 % lower compared to IFSS gotten from the microbond test. A bar chart with the shear strengths and standard deviations is presented in Figure 4. The coefficient of variation (CV) for the microbond results was 5.7 % and for the SBS 9.96 %. The higher CV for SBS can be explained by other fracture modes that might have be present but were not noticed on SEM. rCF is more vulnerable to uneven stress distribution in the SBS since fibers might break in a longitudinal direction during loading. The microbond test ensures comparability between results with identical loading scenarios.

The effect of carbon fiber fabric quality makes comparing different recycled batches challenging with the SBS, as the laminate can have bad areas, where weaving has gaps and distortion. Producing the SBS samples could be impossible if there is not enough good fabric to make the samples. The effect of weaving must also be considered in the SBS, as it is known to affect the distribution of stresses between the matrix and the reinforcement [2]. The microbond test eliminates the effects of weaving quality and stacking sequence of the laminate.

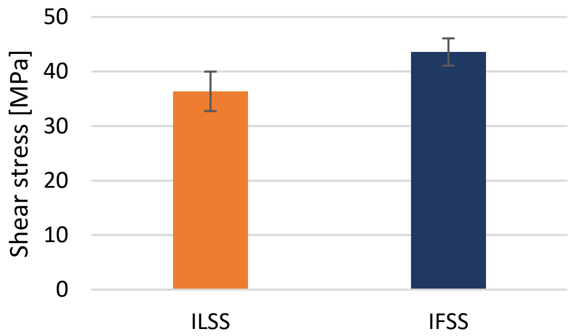

Figure 4. ILSS and IFSS values with standard deviation.

The uneven surface of rCF has a different effect in both tests. In the microbond test, the effect of the uneven surface is emphasized compared to other contributing factors. The uneven surface allows resin droplets to attach to the fiber better through mechanical interlocking, which increases the force required to remove the droplet. The results show to have a good linear fit. The average R^2 value is 0.94. A conclusion about the similar attachment of droplets to the fiber between different samples can be drawn. In the SBS, an uneven surface might increase the likelihood of uneven distribution of the load in the fibers, and thus increases the probability of stress concentrations. The contact stress from the loading points of the SBS will also negatively affect the stress distribution lowering the ILSS and inducing unwanted fracture modes [2].

Stojcevski et al. [11] compared the shear stresses and test methods in the micro-, meso- and macro-scales for carbon fiber and epoxy matrix. The sizing concentrations and the surface treatment of carbon fibers were varied. There were also unsized and carbon fibers. As a micro-scale test, they used a single fiber fragmentation test (SFFS). In their article, they found the shear strength for the unsized fibers to be higher at the macro-scale, than the shear strength of the micro-scale with each surface treatment. Their results differ from the results of this research, where macro-scale results were lower compared to the micro-scale. This is explained by significantly different micro-scale testing methods. The microbond test gives higher results when compared to SFFS. The matrix used was not the same as in this study, but epoxy, nonetheless. The adhesion mechanism can be assumed to be the same for the functional groups on the interface, so a careful comparison can be made between the results.

4. Conclusions

In this study, ILSS and IFSS values were compared, ILSS being 16.6 % lower than IFSS. The microbond test does not consider the volume fraction of fibers, which significantly affects the shear strength on the macro-scale. The fibers carry the load and the more reinforcement there is, the stronger the composite will be. However, SBS sampling is challenging on rCF if there are unevenness and distortion in the fabric. Because of this, the microbond test is a more reliable way to obtain shear strength for a given system when there are no resources in use to verify the quality of the laminate and tested samples and the fabric used is of poor quality. This allows the fiber recycler to compare reliably different manufacturing parameters' effects on shear strength.

Reliability is also increased by many measuring points in the microbond test. The FIBRObond testing device removes the possibility of uneven stress distribution, which produces unwanted fracture modes. In addition, a significant number of droplets from different sizes are tested in the microbond test, allowing the use of reliable linear regression. The results are statistically averaged by the many data points and gives a sufficient view of the fiber's interfacial strength when several fibers are tested from different places of fabric, as was done in this study.

Acknowledgements

This research was partly funded by EU H2020-IND-CE project "FiberEUse" (grant agreement number 730323). This work made use of the Tampere Microscopy Center facilities at Tampere University (33014 Tampere, Finland).

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