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Thermal recycling and re-manufacturing of glass fibre thermosetting composites

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Abstract. The impact of using thermally recycled glass fibre in re-manufactured composites was investigated. A unidirectional glass fibre thermosetting composite laminate was manufactured. The matrix in one part of the laminate was burnt off to recover the glass fibres. These recycled glass fibres were used to manufacture a new composite laminate with the same fibre architecture as the pristine one. The fibres, the matrix and the composite laminates were thoroughly characterised and analysed. The results show that good materials quality was obtained for both laminates. A difference in fibre packing behaviour was observed in the composites with the pristine and the recycled fibres, which lead to a lower fibre volume fraction in the latter one. The Young's modulus of the composites was not changed by the recycling process, if the lower fibre volume fraction is taken into account. However, a marked drop in the maximum stress of the composites was reported, which was found to be related to the loss in maximum stress of the fibres.

1. Introduction

Glass fibre composites are massively used nowadays for their high strength, lightweight and low cost. In 2014, the volume of produced composites was reported to represent 1.04 million tons in Europe [1]. Composites are used in different industry sectors such as transport, electronics, constructions, sports, as well as in the energy sector for wind turbines.

Recycling of wind turbines has become a topic of interest as the first wind farms are currently reaching end of life [2]. The tower, the gearbox, the hub are made of steel, the nacelle is made of aluminium, and these materials have already established recycling routes [3]. However, the blades made of glass fibre reinforced thermosetting polymer composites represent a large volume of the wind turbine, and they are challenging to recycle.

One of the simplest techniques to recycle glass fibre thermosetting composites is to burn off the matrix material and reuse the fibres. The main challenge in this solution is that the properties of burnt glass fibres are dropping with the applied temperature and duration of the recycling process [4,5]. The mechanism for this loss of fibre strength has been intensively studied, and it has been assumed to be due to different aspects, such as surface structural relaxation [6], and water absorption into the glass molecular network leading to formation of defects at the fibre surface [7].

Many studies have characterized the impact of thermal treatments on the properties of glass fibres, however only few have used recycled long continuous glass fibres to manufacture new polymer



composites. Manufacturing of composites with recycled glass fibres is challenging, because the fibres are sensitive to mechanical handling.

The goal of this study is to investigate the effect of using recycled glass fibres on the properties of thermosetting composites. For that, a reference composite made with pristine glass fibres and a composite made with recycled glass fibres are manufactured. In order to ease the evaluation of the results, a simple fibre architecture consisting of unidirectional continuous glass fibres is selected. In addition, the selected manufacturing procedure involves minimal fibre handling in order to minimize damage of the fibres, and to preserve the fibre architecture. In order to understand the impact of the recycled glass fibres on the re-manufactured composite, the properties of the fibre and matrix constituents and the composites are thoroughly characterized and analysed.

2. Materials & Methods

2.1. Materials

Figure 1 presents the experimental procedure applied in this study. First, pristine glass fibres are used to manufacture a composite laminate, which will be named pristine laminate. One part of this laminate is burnt and the glass fibres are recovered. These fibres are named burnt fibres and are used to manufacture a new composite laminate, named re-manufactured laminate. In addition, a sample of pristine glass fibres is burnt at 565°C for testing of fibre properties.

The fibres used are Hybon 2026 PPG glass fibres with a silane sizing type which is dedicated to be used in polyester, vinyl ester and epoxy resin systems. The resin used is a Huntsman 1568 epoxy resin system with Huntsman hardener 3489.

The pristine laminate is a unidirectional fibre composite, which was manufactured using filament winding followed by vacuum assisted resin transfer molding. The laminate was cured in two stages in order to minimize the residual stresses in the matrix. The first stage was performed at 40°C for 19 hours, followed by a second stage at 75°C for 5 hours. The laminate dimensions were 300 x 205 x 2 mm³.

A piece of the pristine laminate with dimensions 150 x 205 mm² was placed on a metal plate in a furnace at 565°C. After 10 hours, the fibres were cooled down to room temperature in the furnace. In order to keep them aligned and avoid direct handling; a second metal plate covered with a release foil was placed on the top of the fibres. The two plates were held together and turned around to have the fibres lying on the infusion plate with the release foil. The fibres were infused and cured with the same conditions as the pristine laminate.

In addition, a pure resin plate was manufactured using the same curing procedure.

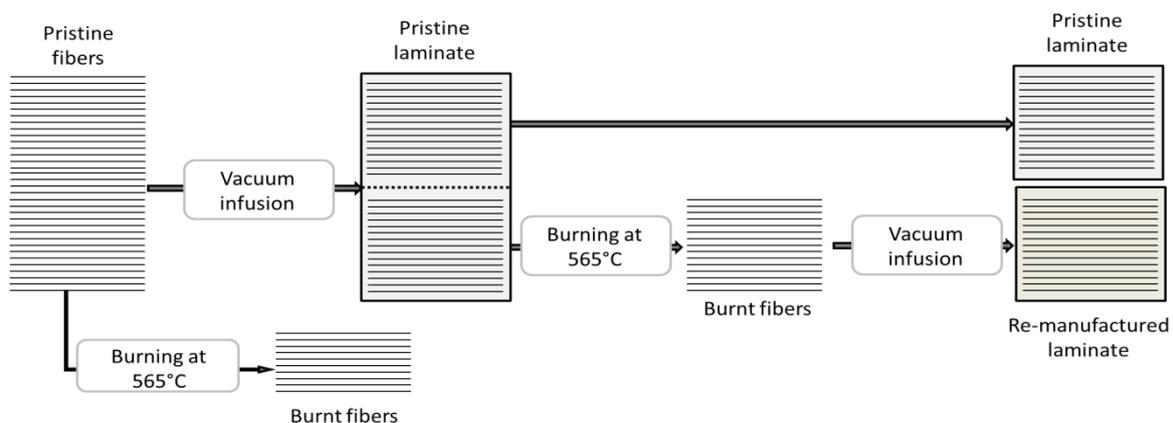


Figure 1. Experimental procedure.

2.2. Fibre characterization

2.2.1 Fibre density. To measure the density of the glass fibres, a gas pycnometer Quantachrome Ultrapyc 1200e was used, with nitrogen as the displacement medium. The fibres were rolled around a spool and placed in the pycnometer.

Before the measurements, the fibre samples (12.0 g of pristine fibres and 9.5 g of burnt fibres) were dried for 24 hours to remove moisture which would influence the measurements in the pycnometer. For both types of fibres, 8 repeated density measurements were performed.

2.2.2 Fibre mechanical properties. Fibres were mechanically tested by an automated single fibre tensile machine (Textechno Favimat+ Robot2, Germany). The test procedure consists firstly in a linear density test, in order to determine the fibre cross-sectional area, and secondly in a tensile test. A 50 mm gauge length was used in order to get convergence during the linear density test. Tensile testing speed was 1 mm/min. For both types of fibres, 50 single fibres were prepared to be tested. The fibre Young's modulus is calculated according to the composite materials standards, between 0.05% and 0.25% strain.

2.3. Matrix characterization

The mechanical properties of the manufactured pure epoxy plate were measured according to the ISO 527-2:1993(E) standard. Six specimens were tested.

2.4. Composite characterization

2.4.1 Volumetric composition. To determine the fibre, matrix and porosity content of the composites, five rectangular samples of dimensions 15 x 15 mm² were cut from each composite laminate. The samples were roughly polished, dried overnight, and weighed. The samples were then sealed with paraffin wax to be able to include the correct porosity volume in the determinations. The sealed samples were weighed first in air, and then in water in order to determine the composite density. Then, the epoxy matrix in the samples was burnt off overnight at 565°C. The weight of the remaining glass fibres was recorded in order to determine the fibre weight content. Using standard equations for composite materials [8], and based on the established values of fibre weight content, composite density, fibre density, and matrix density (set equal to 1.15 g/cm³ [9]), the volume content of fibres, matrix and porosity in the composites was calculated.

2.4.2 Microstructure. To characterize the 2D microstructure of the composites, samples with dimensions 22 x 2 mm² were cut from each laminate. The samples were casted in epoxy, and the sample cross-section in the plane normal to the fibre direction was grinded and polished. Observations were made using an optical microscope.

A Xradia 520 from Zeiss was used to investigate the 3D microstructure of the composites by X-ray tomography. One sample with dimensions 15 x 2 x 2 mm³ was cut from each laminate. To ensure a good scanning quality, the sample width was ensured to be equal to the sample thickness. Since the objective was to describe the composite microstructure, the scans had to cover as large volume as possible, and therefore, scans were stitched together by an image analysis algorithm. The X-ray source was placed at -10 mm and the detector at 25 mm from the sample in order to get a good transmission factor. The accelerating voltage was set to 40 KeV with an exposure time of 5 s using a LE2 filter and 5201 projections to ensure the best quality possible. All these parameters have been chosen to get a voxel size of 1.9 μm³.

2.4.3 Mechanical properties. The mechanical properties of the composites were measured according to the ISO 527-5:2009(E) standard. The length of the tensile specimens was however shortened to 200 mm (instead of 250 mm) as this was the maximum length to fit in the furnace. The tab material used

was 2 mm thick cross-ply glass/epoxy plates produced by Elektro-Isola (Denmark). Seven tensile specimens were tested for each laminate.

2.5. Modelling of composite properties

The composite laminate mechanical properties were analyzed by using the rule of mixtures:

$$E_L = E_f V_f + E_m V_m \quad (1)$$

$$\sigma_L = \sigma_f V_f + \sigma_m^* V_m \quad (2)$$

where E_L , E_f and E_m are the composite laminate, fibre and matrix Young's moduli, respectively, V_f and V_m are the fibre and matrix volume contents, respectively, σ_L and σ_f are the composite laminate and fibre maximum stresses, respectively, and σ_m^* is the stress in the matrix at the composite laminate failure strain.

3. Results and discussion

3.1. Fibres

The density of both pristine and burnt fibres has been measured according to the presented procedure. As shown in Table 1, the pristine fibres have a density of 2.61 g/cm³, and the burnt fibres have a density of 2.64 g/cm³. This increase in fibre density is believed to be due to glass densification during the thermal treatment [10].

Regarding the single fibre testing, 50 fibres of each type were prepared; 42 pristine fibres were successfully tested, while only 17 burnt fibres were successfully tested. This difference is due to that the burnt fibres are brittle, and during clamping of the fibres, a high number of them fail. The measured Young's modulus and maximum stress of the fibres are presented in Table 1. The pristine fibres have a lower Young's modulus than the burnt fibres, with values of 82.2 and 87.5 GPa, respectively. However, the pristine fibres have a much higher maximum stress, 2050 MPa, compared to the maximum stress of the burnt fibres, 400 MPa. Thus, the thermal treatment has impacted the glass fibre maximum stress by a reduction of 80%.

Table 1. Density and mechanical properties of pristine and burnt fibres.

Fibre type	Density (g/cm ³)	Young's modulus (GPa)	Maximum stress (MPa)
Pristine	2.61 ± 0.02	82.2 ± 0.7	2050 ± 520
Burnt	2.64 ± 0.01	87.5 ± 0.8	400 ± 180

Increased stiffness and decreased strength of glass fibres after thermal treatment has also been reported elsewhere in the literature. These phenomena are believed to be due to a densification of the glass molecular network [11] and a removal of sizing. During heating glass fibres in normal atmosphere, the water molecules present in the atmosphere are attacking the fibre surface and replacing atoms. These changes are building stresses at the fibre surface, and this is leading to a decrease of the fibre maximum stress [6].

3.2. Matrix

Based on the measured stress-strain curves of the neat epoxy resin plate, the Young's modulus of the matrix was measured to be equal to 3.1 ± 0.02 GPa, and with a maximum stress of 65.1 ± 0.3 MPa.

3.3. Composites

The two composite laminates were manufactured under the same processing conditions. During manufacturing of the laminate with the recycled fibres, a distinct noise could be heard when the vacuum pressure was applied to the bag with the fibres. This noise is believed to be associated with cracking of the glass fibres, due to their brittleness. A similar noise could also be heard when the resin was infused into the bag with the recycled fibres.

3.3.1 Volumetric composition. Table 2 shows the volumetric composition of the pristine and the re-manufactured laminates. The porosity content was measured to be around 2% for both laminates, which corresponds to a good composite quality. After burning off the matrix in the composite laminates, a small amount of ashes from the combustion was observed on the surface of the fibres. This did not seem to influence the quality of the composites. Regarding the fibre volume content, it was measured to be 59.2% for the pristine laminate and 46.8% for the re-manufactured laminate. Since the same amount of fibres was used for the manufacturing of the two laminates, the difference in fibre volume contents must be due to a difference in the composite volume, which can be assessed by the thickness of the laminates. The average thickness of the pristine laminate was 1.89 mm, while the average thickness of the re-manufactured laminate was 2.21 mm. This indicates that under the same processing conditions, the recycled fibres pack less efficiently than the pristine fibres. To control the laminate thickness and thereby to obtain the same fibre volume content, a frame could be placed around the fibres and a higher pressure could be applied during infusion of the resin.

Table 2. Volumetric composition of the pristine and re-manufactured composite laminates.

Composite laminate type	Fiber volume content (%)	Matrix volume content (%)	Porosity content (%)
Pristine	59.2 ± 0.8	39.1 ± 0.8	1.7 ± 0.1
Re-manufactured	46.8 ± 1.1	51.0 ± 1.1	2.1 ± 0.3

3.3.2 Microstructure. Figure 2 shows optical microscopy images of the microstructure of the pristine and re-manufactured composite laminates. In the pristine laminate, the fibres are well distributed across the laminate thickness, with a few porosities. The microstructure is different in the re-manufactured laminate. The fibres are non-uniformly distributed with large matrix-rich regions. This observation supports the hypothesis that recycled fibres pack less efficiently than pristine fibres. This different fibre packing configuration could be due to the removal of sizing from the fibres during the thermal treatment. One important role of sizing is to avoid electrostatic repulsion between fibres and therefore having a more efficient packing.

Figure 3 shows X-ray tomography images of the microstructure of the re-manufactured composite laminate. The fibres can be seen to be well-aligned, and matrix-rich regions can also be seen, similar to the ones seen by optical microscopy. Careful inspections of the scanned 3D volume revealed a number of locations where the fibres are broken, see 2D projections in Figure 4. These observations support the hypothesis of brittle fibres breaking during manufacturing of the re-manufactured laminate.

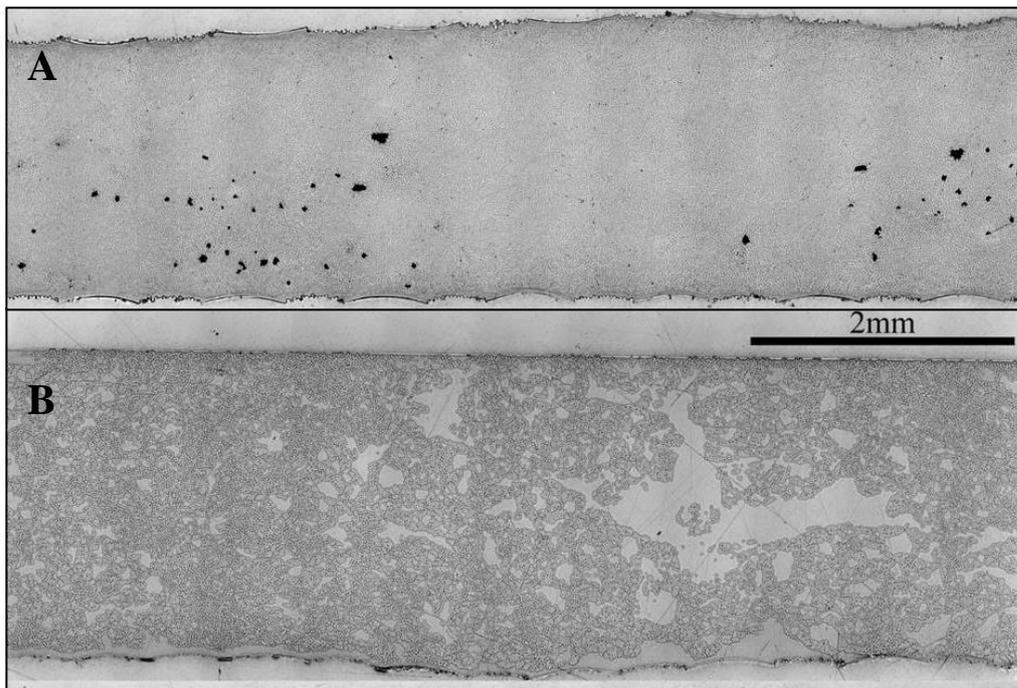


Figure 2. Optical microscopy image of microstructure in (A) pristine composite laminate, and (B) re-manufactured composite laminate.

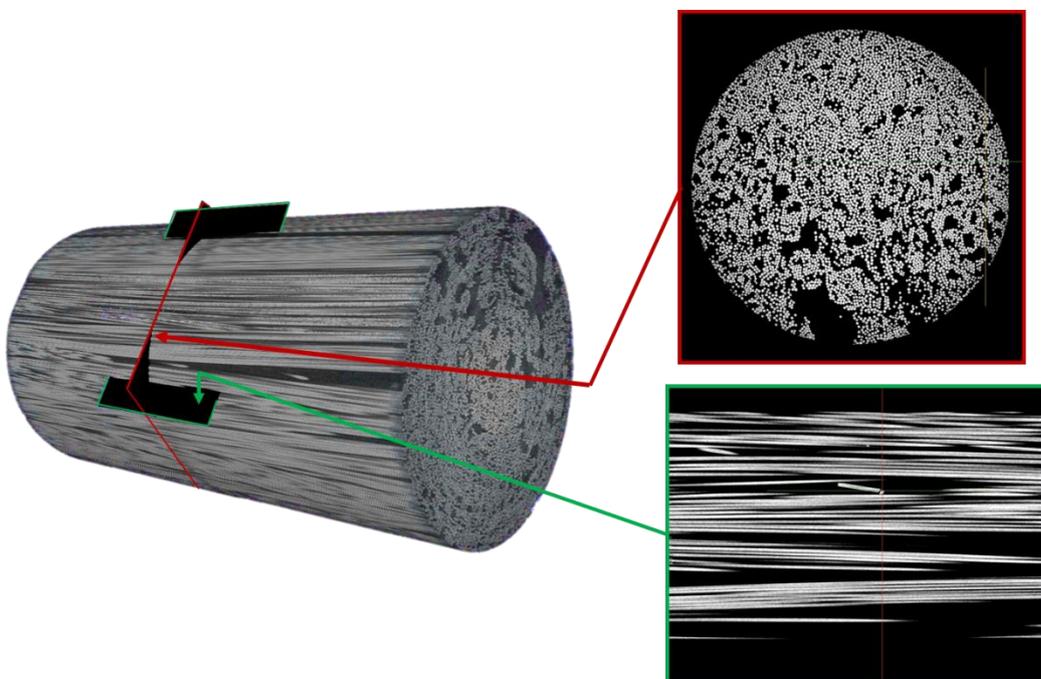


Figure 3. Reconstructed 3D X-ray tomography volume of re-manufactured composite laminate (left), with two examples of 2D projections (right).

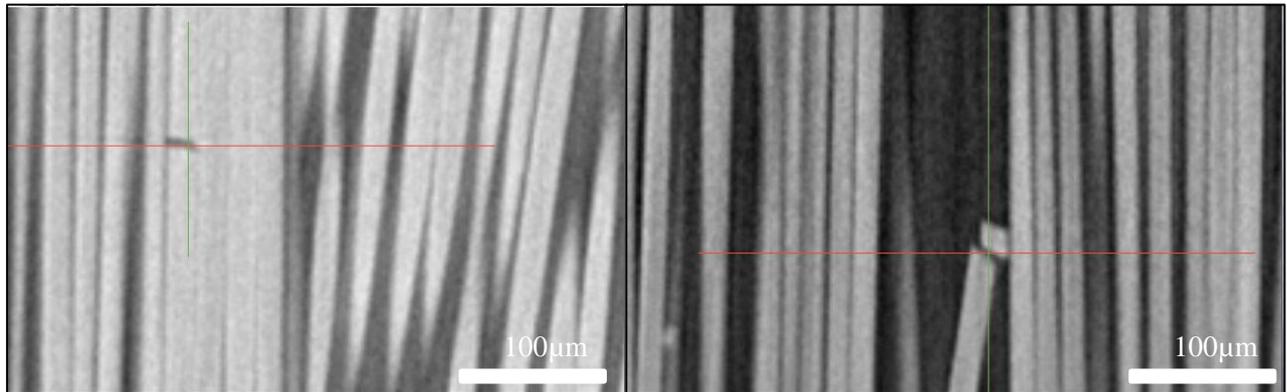


Figure 4. 2D projections of X-ray tomography volume showing broken fibres (marked by crossed lines) in the re-manufactured composite laminate.

3.3.3 Mechanical properties. Results of the tensile tests for the pristine and re-manufactured composite laminate are gathered in Table 3. The pristine laminate has a Young's modulus of 52.8 GPa, whereas the re-manufactured laminate has one of 45.1 GPa. As will be demonstrated later, the main reason for the lower Young's modulus of the re-manufactured laminate is its lower fibre volume fraction (see Table 2). The difference in Young's modulus between the two laminates can also be observed from the stress-strain curves presented in Figure 5, where the curves of the re-manufactured laminate lie slightly under the stress-strain curves of the pristine laminate.

Regarding the maximum stress of the composites, the pristine laminate has a maximum stress of 1210 MPa, whereas the re-manufactured laminate has one of only 120 MPa. This corresponds to a 90% reduction in maximum stress. Static tensile properties of unidirectional composites reflect mostly the fibres properties, and therefore, the low maximum stress of the re-manufactured laminate can be directly related to the low maximum stress of the recycled fibres (see Table 1).

Table 3. Experimental and theoretical mechanical properties of the pristine and re-manufactured composite laminates.

Composite laminate type	Experimental			Theoretical	
	Young's modulus (GPa)	Maximum stress (MPa)	Strain at maximum stress (%)	Young's modulus (GPa)	Maximum stress (MPa)
Pristine	52.8 ± 2.8	1210 ± 70	2.9 ± 0.2	49.9	1240
Re-manufactured	45.1 ± 4.7	120 ± 20	0.3 ± 0.1	42.5	220

The failure behaviour of the tensile specimens can be seen in Figure 6. The specimens from the pristine laminate are showing a chaotic fracture mode with extensive fibre splitting, which is typical for unidirectional glass fibre composites. In contrast, the failure mode of specimens from the re-manufactured laminate is characterised by a single crack across the specimens, and located close to one of the tabs. This difference in failure mode is explained by the difference in maximum stress of the fibres. The pristine fibres are able to carry large stresses before they fail, which then results in a large energy release, leading to a chaotic fracture mode where fibre bundles are split apart in the whole length of the specimens. The recycled fibres are only able to carry small stresses, which does not result in a large energy release at failure, and therefore, the failure mode in these specimens is characterised by a single crack running transverse to the fibres.

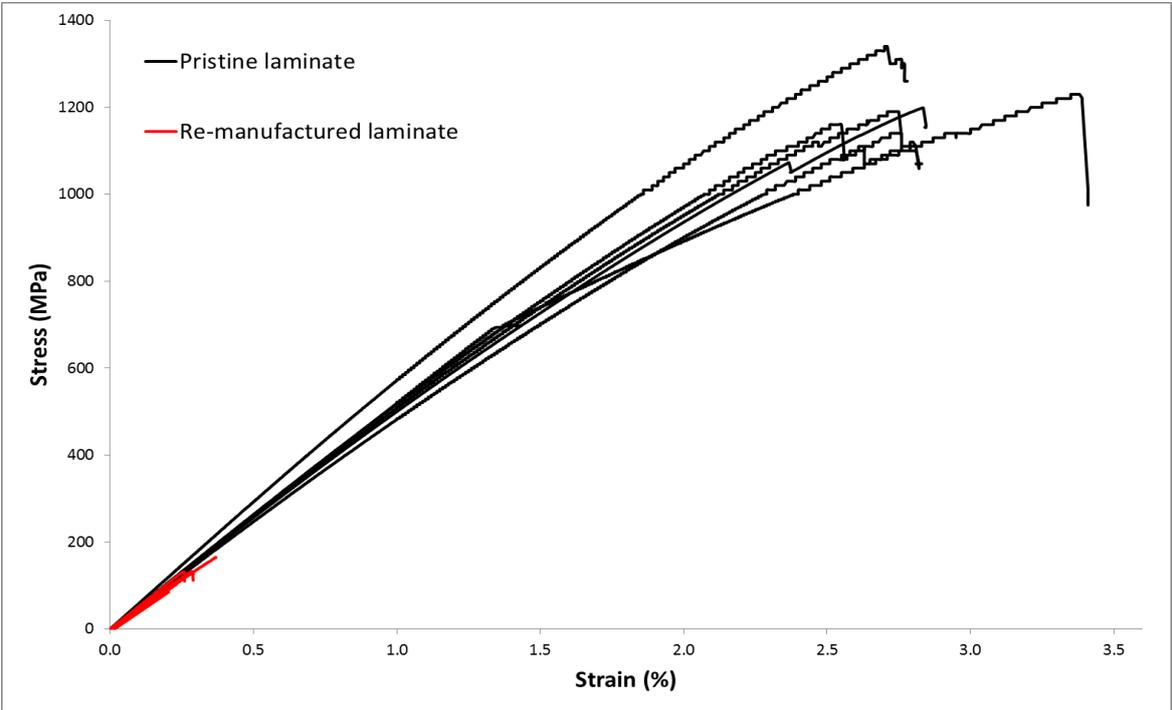


Figure 5. Stress-strain curves of pristine and re-manufactured composite laminates.

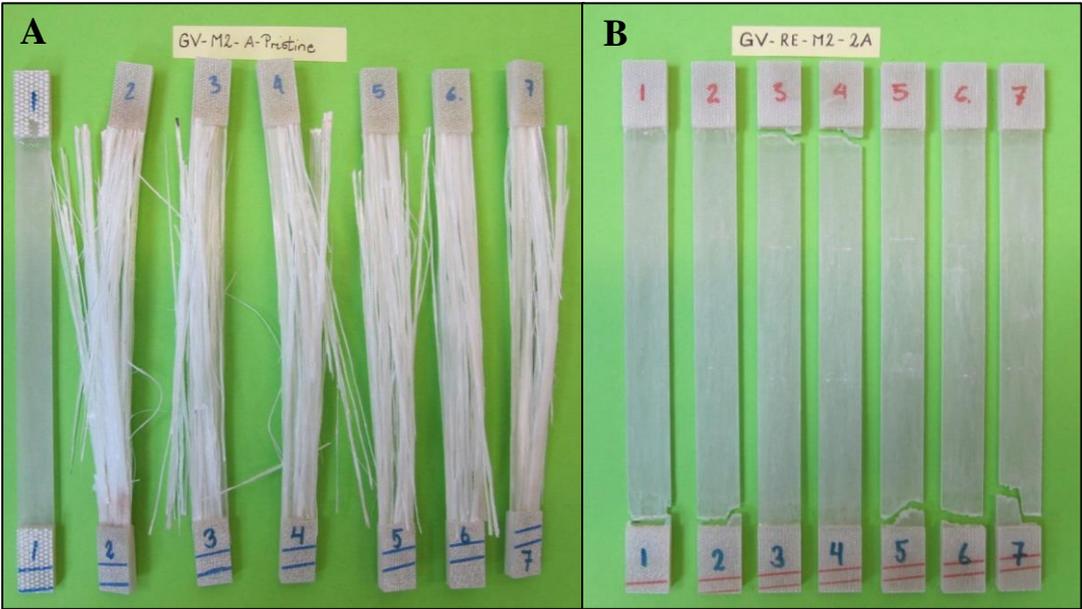


Figure 6. Tensile specimens of (A) pristine and (B) re-manufactured composite laminates after testing.

3.4. Modelling of composite properties

Using the measured volumetric composition of the composites, together with the measured fibre and matrix properties, the theoretical Young's modulus and maximum stress of the pristine and re-manufactured laminates were predicted using the rule of mixtures models, Equations (1) and (2). The results are presented in Table 3.

The theoretical Young's modulus for the pristine laminate is equal to 49.9 GPa, and it is equal to 42.5 GPa for the re-manufactured laminate. As can be observed in Table 3, these results are essentially similar to the measured Young's moduli, taking the scatter (\pm stdv.) of the measurements into account.

The Young's modulus of the re-manufactured laminate can also be estimated by considering it to have the same fibre volume fraction as found in the pristine laminate, i.e. 59.2 %. Then, the theoretical Young's modulus for the re-manufactured laminate is equal to 53.0 GPa. This is similar to the measured Young's modulus of the pristine laminate on 52.8 GPa. It is however larger than the theoretical Young's modulus of the pristine laminate on 49.9 GPa due to the larger Young's modulus of the recycled fibres.

To predict the maximum stress of the composite laminates, the stress in the matrix at composite failure strain was obtained from the measured stress-strain curve of the matrix. This value is equal to 62.8 MPa for the pristine laminate and 8.4 MPa for the re-manufactured laminate. Then, the maximum stress of the pristine and re-manufactured laminates can be predicted to be 1240 and 190 MPa, respectively. Both predictions are in acceptable agreement with the measured values on 1210 and 120 MPa, respectively. The slightly lower measured maximum stress of the re-manufactured laminate can be explained by the failure of these specimens close to the tabs due to stress concentrations. Thus, if more optimal specimen geometry was used (e.g. dog-bone shaped), the measured maximum stress would be expected to approach 190 MPa.

4. Conclusions

Considering optimal manufacturing conditions and fibre alignment, the present study investigates the effect of using recycled glass fibres on the properties of composites. A unidirectional glass fibre thermosetting composite laminate was manufactured, and part of it was burnt to recycle the fibres. The recycled fibres were then used to re-manufacture a new composite laminate using the exact same processing conditions, and involving minimal handling of the fibres in order to avoid damaging them.

The two types of manufactured composite laminates were shown to be of good quality, with low porosity content. The recycled glass fibres could not be packed as efficiently as the pristine fibres, which lead to a lower fibre volume fraction in the re-manufactured laminate. This was assumed to be due to electrostatic attraction between the recycled fibres where the sizing is removed.

Regarding the performance of the composites, the Young's modulus was not significantly impacted by the recycling process. The recycled glass fibres were found to have a larger Young's modulus than the pristine glass fibres, which theoretically also would lead to a larger Young's modulus of the re-manufactured laminate, in case of similar fibre volume fractions. The maximum stress of the re-manufactured laminate showed a 90% reduction compared to the pristine laminate. This can be attributed to the lower maximum stress of the recycled glass fibres.

Regarding the impact of the manufacturing process on the re-manufactured laminate, it was not possible to avoid it completely. It was shown that the brittle recycled fibres were breaking during the vacuum infusion process.

The work in the present study does not involve measurements of the possible changed adhesion of the recycled glass fibres to the thermosetting matrix. Further studies should be focused on characterizing the fibre/matrix adhesion in re-manufactured glass fibre composites.

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