A COMPARATIVE STUDY INTO MECHANICAL PERFORMANCE OF GLASS FIBRES RECOVERED THROUGH MECHANICAL GRINDING AND HIGH VOLTAGE PULSE POWER FRAGMENTATION

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Abstract

This study investigates the effectiveness of the High Voltage pulse power fragmentation and the quality of its recyclates, in comparison to the well-established mechanical grinding process. Short glass fibre reinforced polyester resin composite panels were produced to simulate typical composite waste streams. These panels were then subjected to both mechanical grinding and high voltage pulse power fragmentation processes. Recyclates were collected for further morphological analysis and glass fibres extracted for detailed observation of surface features and measurements of resin content. In order to confirm the quality of recycled glass fibres and their suitability for further reuse, mechanical properties of the fibres were carefully measured, and fibre length distribution was also compared.

Introduction

Glass fibre reinforced composites have been widely used in a broad range of applications in automotive, aerospace and construction industries.[1, 2] These composites are comprised of glass fibres and resin. In some cases they also contain fillers which are typically cheap mineral powders. The proportion and type of resin, reinforcement material and fillers can be varied and tailored according to the specific application.

Sheet Moulding Compounds (SMC) have been widely used as the most successful automotive structure. The compression moulding of SMCs has been introduced as a suitable technique to manufacture large volumes of composites at a low cost, sufficient mechanical properties and a good surface finish. SMCs are typically composed of 25 mm randomly distributed glass fibre bundles as the reinforcing material, polyester resin and other components such as filler, catalyst, pigment and other additives.[2, 3]

Approximately, one million tonnes of composites are produced every year.[4] Composite recycling is a challenge for many industries as the use of composites for various applications increases.[5] The continuous rise in demand for lightweight structures, combined with the increased pressure of legislation and landfill cost, has resulted in the need for recycling of thermoset composites from off-cuts during manufacture and end-of-life components. Several recycling technologies have been proposed for the recovery and reuse of thermoset composites. Mechanical recycling, solvolysis, pyrolysis and fluidised bed are some of these recycling techniques, investigated thoroughly throughout the literature.[6]

Mechanical grinding is a low cost and straightforward process to break up composite wastes. However, the grinding process can result in a significant loss in the quality of recyclates, especially of the fibrous reinforcement component.[6-8] The majority of previous studies using this technique typically reduce the size of the scrap material down to a fine filler using a high speed mill.[7, 9, 10] Only a few studies carefully controlled the grinding process to allow the recovery of short glass fibres for reuse as reinforcement.[1, 11] However, the mechanical characterisation of the recovered fibres showed deterioration in strength.[1, 11]

A novel approach based on High Voltage (HV) pulse power fragmentation which was originally used for mining applications, has been developed for breaking down composite structures. Electrical discharges with a pulse power range of 50 kV-200 kV are applied in an aqueous environment. Shocking energy can be transferred into the solid material that can cause detachment along the internal material boundaries. This ultimately results in a relatively selective liberation of the structural components of the composites.[12]

The main aim of this work is to study the HV pulse power fragmentation as a novel approach for recovery of glass fibres by comparison with mechanical grinding. SMC panels were fabricated. Mechanical grinding and HV pulse power fragmentation were then used to recover glass fibres from the composite panels. Morphology study and single fibre test were carried out on the glass fibres. The quality of the recovered fibres was investigated, and their mechanical properties were measured and compared. Fibre length distribution and resin content on the recycled fibres were also determined for the two recycling processes.

1. Experimental

1.1. SMC production

SMCs were manufactured on a lab scale SMC line at the University of Exeter. Batches of raw composites with a total weight of 12 Kg (sufficient to produce 10-12 SMC panels) were prepared according to a typical SMC formulation used in an automotive production line. SMCs were fabricated using MultiStar ® 254 glass fibres as received from Johns Manville. SMC production is a continuous manufacturing process where measured quantities of chopped glass fibres are delivered evenly onto a moving bed of SMC paste. Once compounded, the formulation was allowed to mature at 30 ºC for 48 hours. Compression moulding was subsequently performed using a hot press. The formulation was cured in the hot press at 145 ºC and with pressing force of 220 kN for 4 minutes. Flat SMC panels with dimensions of 220 × 220 × 4 mm were moulded using this process.

1.2. Mechanical grinding

After an initial size reduction, the SMC panels were fed into an Alpine UPZ-II hammer mill (Hosokawa Micron Ltd, UK) with 25 mm classifier screens. The recovered glass fibres are shown and compared to virgin fibres in figure 1.

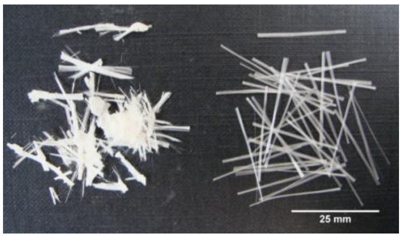


Figure 1. Mechanical ground fibres in comparison with virgin fibres

1.3. HV pulse power fragmentation

Another group of SMC panels were reduced in size and then subjected to the HV pulse power fragmentation process (SELFRAG AG, Switzerland).[13] The SMC panels were cut into sections of 8 × 4 cm size using a band saw. The composite pieces were then placed in water. Water acts as a medium with the required electrical properties which is accessible, nontoxic, cheap and can be easily removed from the target material.[14] A pulsed electrical discharge with 1-5 ms and energy of 10-100 $J \times cm^{-1}$ is guided through the composite material at temperatures above 975 ºC and pressures of 10^9-10^{10} Pa.[14, 15] This process is also called electro-dynamic fragmentation. The mechanism of the process is shown in figure 2. As it is presented in figure 2, a voltage pulse of 150 kV, with a fast pulse rise time is produced between the two electrodes. A discharge occurs through the composite material (the dielectric strength of the material needs to be lower than that of ambient). A schematic image of the fragmentation process is shown in figure 2.

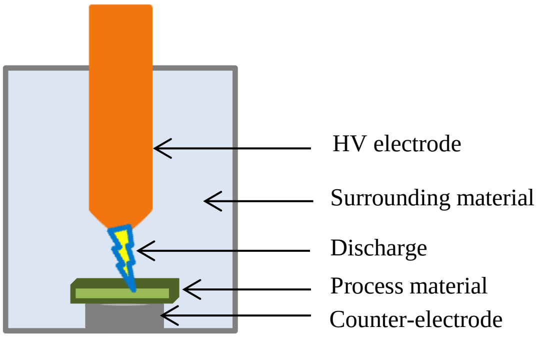


Figure 2. Schematic image of the HV pulse power fragmentation process

In the HV pulse power fragmentation process, the electrical discharge diffuses into the composite material and weakens the material along the boundaries, which results in formation of cracks. A shock wave is then generated as the deposited energy propagates into the material and compressive stress on the material increases. This causes breakages at the boundaries and weak interfaces, and results in the material breaking into its components.[14, 15] The process is described in detail in a study by Bluhm *et al*.[15] The recovered fibres are presented and compared to virgin fibres in figure 3.

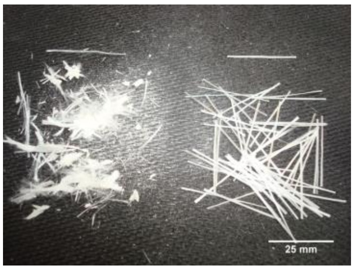


Figure 3. HV fragmented fibres in comparison with virgin fibres

2. Testing

2.1. Heat treatment on the recycled glass fibres

The heat treatment process was carried out on the recycled glass fibres from both recycling technologies in order to determine the resin content on the fibres. The fibres were weighed before the heat treatment using a Mettler-Toledo XS 205 dual range analytical balance. The heat treatment process performed on the fibres included a gradual increase in temperature to prevent fibre breakages during the ashing process as a result of high temperature:

a) Heat from 30 °C to 300 °C, maintain sample at 300 °C for 2 hours
b) Maintain the samples at 300 °C for 30 minutes
c) Heat sample from 300 °C to 650 °C at 1.16 °C × min^{-1}
d) Maintain the temperature at 650 °C for 40 minutes
e) Allow the natural cool down of the sample

As a result of the heating process, the residual resin on the fibres disintegrated. However, the resin was still present on the surface. In order to remove the resin, the fibres were washed using water and dried in an oven at 40 °C after the heat treatment. This resulted in clean fibres which were then weighed. The resin content was measured as the difference in the weight values of the recycled glass fibres before and after the ashing process. The ashing process was carried out on six different bundles for each recycling technique. The average value was then calculated for the fibres for each technique.

2.2. Fibre morphology

The surface of virgin and recycled glass fibres were examined using SEM. The fibres were gold coated to avoid the increasing amount of surface charge. The fibres were assessed using an Hitachi S-3200N SEM.

2.3. *Fibre length distribution*

The fibre length distributions of the ‘as recovered’ fibres (not ashed) were determined using a digital camera and ImageJ software. This was carried out for both the mechanical ground and HV pulse power fragmented glass fibres. 20 images of various groups of fibres were captured for each type of recovered fibres, and each image contained around 30 fibres.

2.4. Micro-tensile testing

Mechanical tests were carried out on virgin and recycled (both mechanical grinding and HV pulse power fragmentation) glass fibres. The sample preparation and mechanical characterisation were performed on single glass fibres according to ASTM D3379-75 standard test method. A single straight glass fibre was mounted and centred on a window card with a 5 mm gauge length in the middle. The fibre ends were glued to the two sides of the window using cyanoacrylate adhesive. The edges of the window card were cut after mounting the specimen on a single fibre deformation rig with 10 N load cell and precision Linear Variable Differential Transformer (LVDT) driving system. A schematic figure of the tensile testing rig is presented in a study by Kao *et al.*[16]

The diameters of all the fibres (fibres considered for the mechanical testing) were measured using an optical microscope prior to the tensile test. The fibres were tested at 1 mm×min^{-1} cross head speed. The mechanical tests were performed on more than 50 specimens for each type of the glass fibres. Tensile strength, Young's modulus and failure strain of the single fibres were measured and compared.

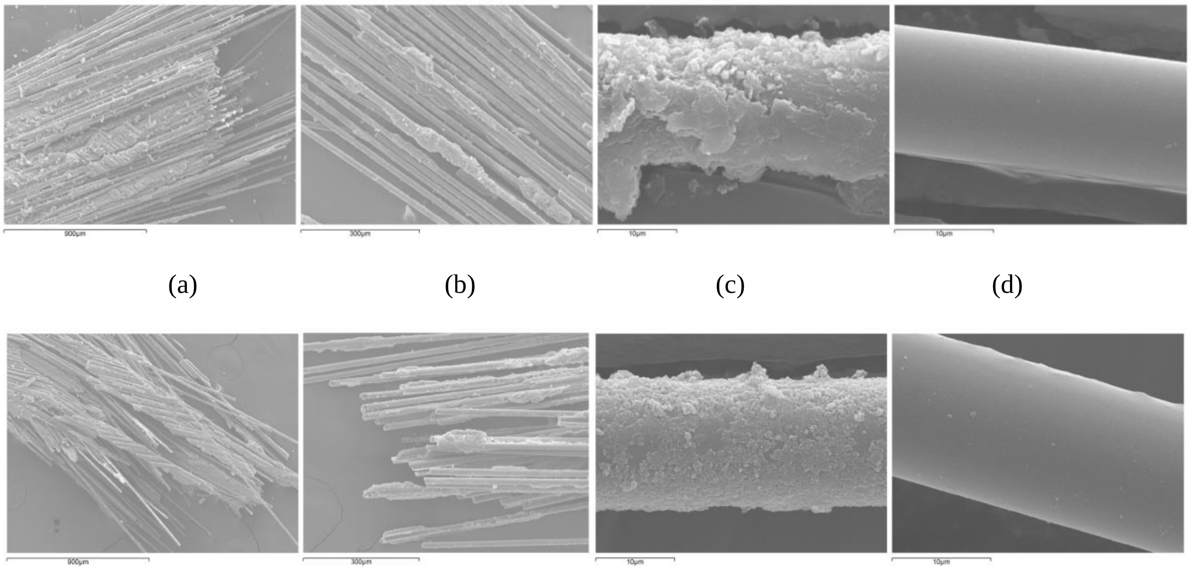
3. Results

3.1. Resin content

The ashed glass fibres were clean and free of residual resin (SEM results presented later confirm this.). However, the fibres were very weak and brittle. The glass fibres have been reported to be sensitive to heat treatments.[17-20] Based on the weight of the fibres before and after the ashing process, the resin content was calculated as 54% by weight for the mechanical ground fibres and 45% by weight for the HV fragmented fibres. This indicates a lower resin content for the fibres recovered from the HV fragmentation process.

3.2. SEM results

SEM images of the recycled fibres were obtained with various magnifications. These are shown in figure 4.



(a) (b) (c) (d)

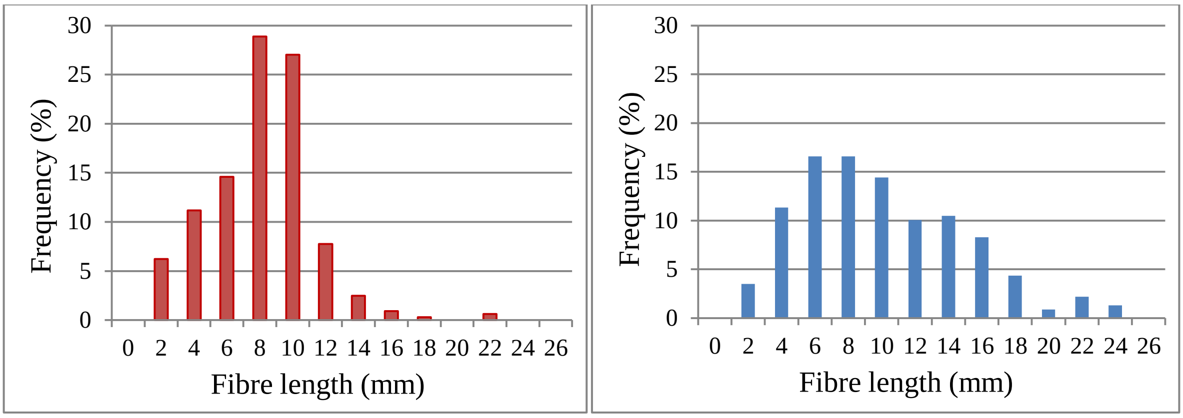
(e) (f) (g) (h)

Figure 4. SEM images of the recovered glass fibre bundles with various magnifications: (a) mechanical ground fibres ×18, (b) mechanical ground fibres ×45, (c) mechanical ground fibres ×900, (d) mechanical ground fibres ×1100, (e) HV pulse power fragmentation ×18, (f) HV fragmentation ×43, (g) HV fragmentation ×900, (h) HV fragmentation ×1100

As it can be seen in figure 4, the majority of the recovered fibres from both recycling techniques are covered with residual resin at the surface. The polyester resin appeared partially removed and remained on the fibres. Grains of recyclate can also be seen on the surface of the fibres. Similar SEM results were obtained by Palmer *et al*. that used mechanical grinding as the recycling process of SMC panels, and Roux *et al*. that used HV pulse power fragmentation to break fibre reinforced polymer structures.[1, 12] Depending on the recycling process used, the amount of residual resin on the surface of the fibres can vary significantly. For example previous studies based on hydrolysis showed an average 15% (by weight) resin content left on of the surface of the glass fibres.[16]

3.3. Fibre length distribution results

The fibre length distributions for both the mechanical ground and HV fragmented glass fibres were produced. The length distributions are presented in figure 5.



(a) (b)

Figure 5. Fibre length distributions for the recycled fibres from different recycling technologies: (a) Mechanical grinding, (b) HV pulse power fragmentation

Considering the fact that glass fibres with 25 mm lengths were used to fabricate the SMCs, a loss in fibre length can be observed after the recycling process in both cases (figures 5a and 5b). ERCOM in Germany used a hammer mill to break SMC panels, and measured a fibre length of up to 20 mm for the recovered glass fibres.[10] Palmer *et al*. used mechanical grinding, and recovered glass fibres with a length of 8-25 mm.[1] In this study, a wider length distribution was measured for the HV fragmented glass fibres (figure 5b) in comparison with that of mechanical ground fibres (figure 5a). In the case of mechanical ground fibres, the majority of the fibres are in the range of 2-16 mm with the highest frequency at 8-10 mm. However, the fibre spread for HV fragmented fibres is 4-20 mm.

3.4. Mechanical properties of the fibres

The measured tensile strength, Young’s modulus and failure strain of the single virgin and recovered glass fibres are presented in Table 1. The diameters of the fibres were determined using optical microscope images prior to the tensile testing and are indicated in Table 1. The tensile strength, Young’s modulus and failure strain showed lower values for the recovered glass fibres compared to the virgin ones. The HV fragmented and mechanical ground glass fibres presented 11% and 19% reduction in tensile strength respectively, and 12% and 38% drop in Young’s modulus respectively, in comparison with virgin fibres. The failure strain also showed a decrease for the recovered fibres. A higher tensile strength and Young’s modulus were obtained for the HV fragmented glass fibres compared to the mechanical ground fibres, with approximately similar value of failure strain in both cases. HV fragmentation introduces some advantages in comparison with a conventional grinding process, including less reduction in mechanical performance of glass fibres, no direct mechanical contact and significant reduction of contamination during HV fragmentation (due to the lower resin content) [14]. The HV process could potentially be further improved to allow separation of even cleaner fibres with less damage to mechanical performance [14]. It is also possible to vary the deposited energy over large areas for the HV fragmentation technique. This can provide the opportunity to separate one or more components of the composite structure as intact as possible and at maximum sizes, and therefore leads to longer glass fibres with better mechanical properties.[14]

Fibre	**Tensile strength (GPa)**	**Young's modulus (GPa)**	**Failure strain (%)**	**Diameter (µm)**
Virgin	2.14±0.81	63.48±26.68	6.73±2.81	15.91±1.73
HV fragmented	1.89±0.53	55.65±15.83	3.98±1.67	15.57±1.09
Mechanical ground	1.73±0.66	39.1±17.78	3.96±1.75	15.58±1.68

Table 1. Mechanical properties and diameters of virgin and recovered glass fibres from the HV fragmentation and mechanical grinding processes

These mechanical properties were previously measured by Palmer *et al*. (2009) for virgin and mechanical ground glass fibres at 5 mm gauge length. The tensile strength measured by Palmer *et al*. for virgin and mechanical ground fibres is comparable with that measured in this study. The Young's modulus measured in Palmer's study for virgin fibres is similar to that of the current study. However, lower Young's modulus was determined for the mechanical ground fibres in this study compared to Palmer's study. Failure strain of both virgin and mechanical ground fibres are higher in this study compared to those of Palmer's study.[11] The reasons for the different modulus and failure strain obtained in this study and Palmer's study may be due to the fact that a different mechanical grinding machine has been used in Palmer's study, and the amount of residual resin left on the fibres might be different. This could ultimately lead to earlier breakage and lower strain and modulus, with the resin acting as crack initiator or stress concentrator.

Conclusions

The SMC panels were subjected to the novel HV pulse power fragmentation technique as well as the mechanical grinding process. The residual resin contents and fibre length distributions were measured for the recovered glass fibres. The surface morphology and mechanical properties of the recovered fibres were measured and compared with those of virgin glass fibres. Lower amounts of residual resin and a wider fibre length distribution were measured for the HV fragmented fibres, in comparison with the mechanical ground fibres. The surface characterisation showed that glass fibres are unevenly covered with resin. Clean fibres were obtained by subjecting the recovered fibres to the ashing process. A good preservation of mechanical properties was observed for both HV fragmented and mechanical ground fibres compared to the virgin fibres. The tensile strength and Young's modulus measured for the fibres recovered from the HV fragmentation process were slightly higher than those recovered from the mechanical grinding process. However, the improvement in strength is not significantly higher to justify the use of the HV fragmentation technology over the grinding methods. In addition, the throughput for composite recovery from the two processes is significantly different; the current HV fragmentation system is not optimised for composite recycling and therefore the rate of composite recycling is significantly lower.

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