RECYCLATE FIBRES –MATRIX INTERFACE ANALYSIS FOR REUSE IN SHEET MOULDING COMPOUNDS (SMC)

James Palmer[1], Oana R. Ghita[1], Luke Savage[1], and Ken E. Evans[1]

[1]School of Engineering, Computing and Mathematics, Exeter University, North Park Road, Exeter, EX4 4QF, UK

ABSTRACT

In order to optimize and enhance the mechanical properties of sheet and dough molding compounds (SMCs and DMCs respectively) incorporating recycled materials, this study investigates the behaviour of individual recycle components (fibre, resin and filler-$CaCO_3$) within virgin polyester matrix. Hence, recycle glass fibre-polyester resin interfaces are investigated and compared with standard virgin glass fibre-polyester resin using single tensile tests and pull-out tests. Similarly, the bonding interfaces of resin and $CaCO_3$ recycle filler components within virgin polyester resin are characterized using scanning electron microscopy (SEM). The recycle used for investigation was obtained through grinding processes and separated in several fractions using air and sieving classification. The results revealed that the deterioration of the fibre during grinding leads to a decrease in strength of recycle fibres, while the SMC residue left on them significantly reduces the fibre-matrix bond strength.

1. INTRODUCTION

This study investigates the potential of reusing SMC composite recycle (obtained through mechanical recycling) as reinforcement in new moulding compounds, focusing on the interfacial analysis of recycle components with a new polyester resin matrix.

Previous studies [1] concluded that composites incorporating regrind tend to exhibit significantly inferior mechanical properties but no investigations into the reasons behind this behaviour have so far been conducted. The decrease in mechanical properties is not surprising considering that in most cases the recycle is ground to fine particles and reintroduced into virgin structures as filler replacing $CaCO_3$. The typical particle size of ground recycle used in this way vary from 1-5mm [2] to approximately 40μm [3, 4], hence losing a significant part of the reinforcement. In the past, manufacturing of composites incorporating composite recycle has been carried out in a relatively simplistic manner [3, 5] without the requisite background research into the characteristics of composite regrind (fibre length, structure-content of regrind) and without an in-depth understanding of the mechanical properties of the interface between virgin and recycled materials.It is well established that the mechanical properties of the matrix-matrix and matrix-fibre interfaces are crucial as they dictate the mechanical behaviour of the overall composite structure. Only two previous studies recognised the importance of understanding the mechanical properties of recycle fibres [6] or the significance of recycle glass fibre-matrix bonding [7] within a composite structure. The strength of glass fibres recovered through a fluidised bed process [6] at temperature varying from 450 to 650°C, was investigated prior to being used in new DMC structures. The study showed a decrease in strength of approximately 50% due to the effect of high temperature exposure.

One previous study [7] investigated the chemical treatment of the recycle fibre, in order to enhance the interface bonding between recycle glass fibres obtained through regrind and polyester matrix. Although the surface treatment investigated in the study

[8] did not show any improvement on the mechanical performance of the new composites, this is possibly the only study focused on the fundamental understanding of recyclate fibre characterisation, fibre length and analysis of recyclate.
This paper focuses on analysis of the interfacial properties within composites containing recyclate material by determining the behaviour of the individual components (recyclate resin particles, and fibres) within a virgin polyester matrix. In the past, several papers [8, 9] discussed the interfacial behaviour of virgin glass fibres embedded in various polyester matrices using the pull-out method with mechanical or energy models. Although it is recognized as one of the most difficult test methods to be carried out, the pull-out test is the most employed method as (i) it provides direct measurement of debonding force vs. embedded length; (ii) allows calculation of several interfacial parameters and (iii) simulates one of the processes of failure in reinforced composites. For these reasons, pull-out test has been employed in the present study.
Although the interfacial parameters resulting from the use of various fibre coatings and fibre sizing embedded in different matrices has been extensively investigated [9, 10, 11], no attempt has yet been made to investigate the strength of recyclate glass fibres obtained through regrind and the nature of the interfacial bond when used in reinforced composites.

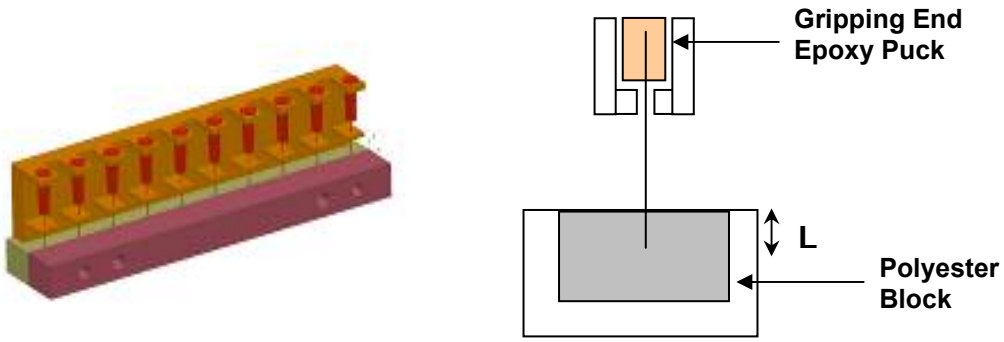
2. EXPERIMENTS

2.1 Fibre preparation

Production-waste automotive front fender sections made of SMC were used as the source of recyclate materials. These were cut into 20 x 20 cm pieces and then granulated using a screen-classifier type rotating hammer, to produce the long fibre recyclate material required for this investigation. Virgin E-glass fibres were supplied by the SMC manufacturer for comparison, and all individual test fibres were carefully removed from fibre bundles immediately prior to sample preparation.
The average fibre diameter was measured as 16μm for all fibres, using SEM, and although the images show that the recyclate materials have areas of resin coating, (Figure 3), this did not affect the average fibre diameter of the specimens measured.

2.2 Pull –out test

Pull-out test sample preparation was performed in two stages:
(1) To allow more accurate handling of the individual fibres, they were initially set in small epoxy pucks (EPOTEK 375). The fibre was clamped through the epoxy puck in the jig and the free end of the fibre was embedded in polyester resin, to a controlled depth, (Figure 1b). The curing profile of the polyester resin blocks took place in two stages: (i) 90 to 120°C for 3h, and (ii) post-curing for 1h at 150°C. The sample was removed from the jig, the epoxy puck cut off and the free fibre length was measured using a shadow micrograph.
(2) Finally, a small epoxy tab (EPOTEK) was cured to the free fibre length, as close as possible to the polyester block, to make in easier to grip in the test machine.



(a) (b)

Figure 1: (a) Pull-out jig design (b) Individual pull-out set-up

2.3 Micro-tensile testing

Single fibres were carefully removed from the virgin and recyclate bundles, minimising the creation of further defects, and samples prepared in accordance with ASTM D3379 'Standard test method for tensile strength and Young's modulus for high-modulus single-filament materials'. The tensile testing was performed using a Biax-200 Micromaterials, with a 10N load cell, at a crosshead speed of 5% strain rate for each gauge length, the extension of the fibre was measured by a linear variable displacement transducer (LDVT). The load cell and LDVT were interfaced with a computer, and the tension and extension of the samples were recorded during the tests.

Individual fibres were mounted on pre-cut cardboard mountings (see Figure 2) using a fast drying cyanoacrylate adhesive. The cardboard mounted samples were then placed thus that the sample was aligned axially between the jaws of the testing machine, and then gripped in place. Both sides of the cardboard mounting were cut to leave just the glass fibre suspended between the grips, so that only the fibre is loaded during the test. Tests were run until the fibre failed, all samples exhibited a linear force-deflection response until a brittle failure. All samples that failed at the grips were discarded. 50+ samples for each of the three gauge lengths (5mm, 10mm and 15mm) for both virgin and recyclate fibres were collected. (The maximum gauge length was limited by the reduced fibre lengths after granulation of the SMC).

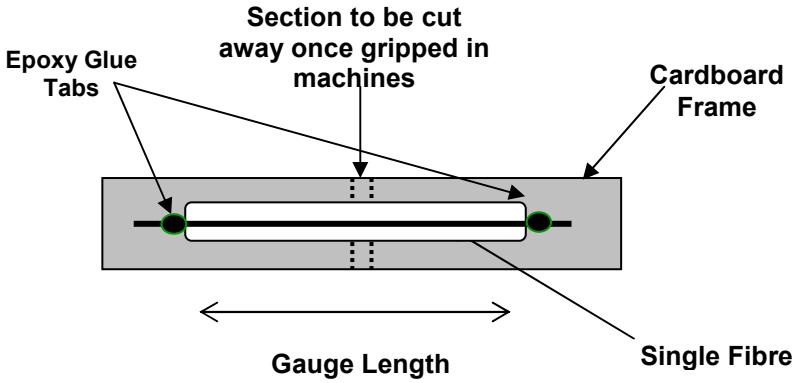


Figure 2: Micro-tensile test set-up

2.4 Fracture Surface Analysis -SEM

In order to understand the nature of the bonding between individual components of SMC recyclate with the virgin resin, filled polyester resin bars were produced and notched and then broken so that their fractured surfaces could be examined using SEM. A range of particle sizes were examined using three different filler materials:

(i) Cured polyester resin powder (self reinforced)
(ii) $CaCO_3$ powder
(iii) Recyclate powder

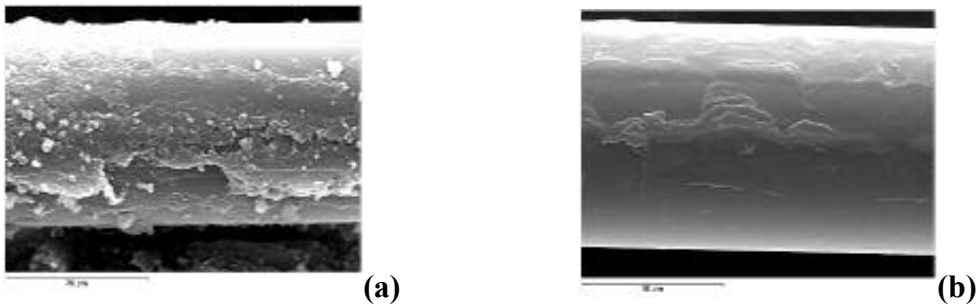
Each of these fillers were classified by dry sieving with mesh sizes 20 to 250 and used to produce 10x10x80 mm polyester filled samples in a specially made mold, at 10% filler by volume.

The SEM examinations of the materials and samples used and tested in this investigation were performed using a Hitachi S-3200N scanning electron microscope. All samples were given a 4nm gold coating to reduce surface charging and secondary electron images were taken with an accelerating voltage of 25kV.

4. EXPERIMENTAL RESULTS AND DISCUSSION

4.1 Single fibre pull-out test

A high magnification SEM analysis of recyclate fibres showed that they retained a significant amount of SMC residue on the surface while virgin fibres are manufactured with a thin silane coating layer, as mentioned in previous investigations. Figure 3 gives an example of the morphology of the recyclate and virgin fibre coatings.



(a) **(b)**

Figure 3: SEM of (a) recyclate and (b) virgin glass fibres

This is the first indication that the mechanical properties of recyclate fibres are likely to be significantly different than those of the virgin fibres. Although the SMC paste retained on the recyclate fibres could act as new interface and increased surface area for bonding with the matrix, the fact that the initial silane coating on the virgin fibres has been covered\reacted and the reported poor thermoset-thermoset resin bonding suggest that this will actually reduced the strength of the resin-fibre interface.

Figure 4 reports the maximum debond load versus embedded length for virgin and recyclate fibres.

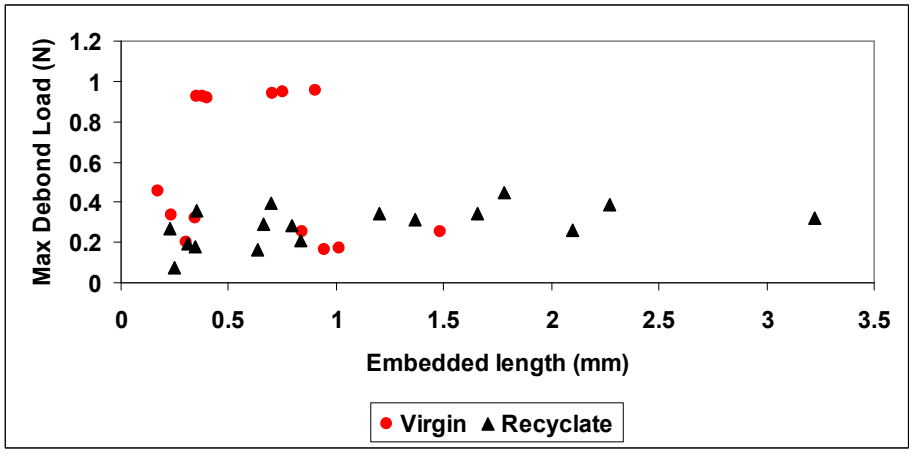


Figure 4: Maximum Debond Load versus embedded length for virgin and recyclate fibres.

In comparison with the conventional pull-out data where the load necessary to debond and pull-out increases with fibre embedded length, it is apparent from Figure 4 that in the case of recyclate fibres, the debond force remained within the same load range (0.2-0.5N) independent of the embedded length. Pull-out of recyclate fibres was possible up to 2.5mm embedded length, whereas the virgin fibre samples failed in the fibre before pull-out at embedded lengths higher than 1mm; hence the lack of data on the virgin fibres at longer embedded length. Although more tests are required in the case of virgin fibres to better highlight the trend, it can be seen that there is a rapid increase in maximum debond load of virgin fibres up to approximately 1mm embedded length at which fibre failure was observed.

The average shear stress was calculated using the mechanical approach based on Lawrence's model [9], where the interface fails when the interfacial shear stress exceeds the debond shear strength (τ_d).

$$\tau_d = \frac{F_d}{2\pi r_f L} = \tau_d \frac{\tanh(\alpha L)}{\alpha L} \qquad (1)$$

Where F_d is the debond load, r_f is the fibre radius, α is an elastic constant and L represents the embedded length.

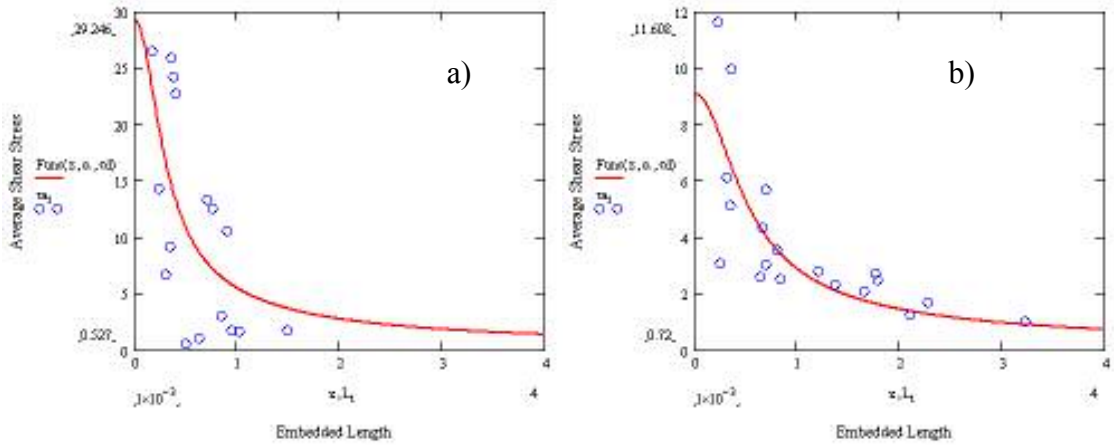


Figure 5: Average Shear Stress vs. embedded length (a) virgin and (b) recyclate glass fibres (Experimental o and Model —).

Although a good agreement between theoretical and experimental values was obtained for both types of fibres, there was a significant drop in the debond shear strength of the recyclate fibre from approximately 30MPa to 12MPa.

4.2 Single fibre tensile test

The elongation at break, ultimate strength and elastic modulus of virgin and recyclate glass fibres at 5, 10, 15mm gauge lengths is detailed in Table 1.

Fibre	**Gauge length (mm)**		
Virgin Fibre	**5**	**10**	**15**
Elongation at break (%)	3.6 ± 0.01	3.1 ± 0.01	3.1 ± 0.006
Ultimate strength (GPa)	2.08 ± 0.6	2.09 ± 0.6	2.12 ± 0.6
Modulus (GPa)	53.8 ± 10	67.8 ± 9.2	69.3 ± 12.8
Recyclate Fibre			
Elongation at break (%)	2.8 ± 0.01	2.6 ± 0.007	2.3 ± 0.007
Ultimate strength (GPa)	1.72 ± 0.6	1.64 ± 0.5	1.48 ± 0.5
Modulus (GPa)	62.1 ± 12	62.7 ± 11	63.7 ± 9.8

Table 1 Properties of virgin and recyclate fibres tensile tested

Results in Table 1 shows that there is a significant decrease in properties of recyclate fibres. The drop in elongation at break between virgin and recyclate fibres could be associated with the two different types of coating present on the fibres. It is envisaged that the silane coating, being more ductile, will stop cracks propagation and grows, where the uneven, brittle resin coating of the recyclate fibres will not give the same effect. In contrast, the drop in ultimate strength of recyclate fibres could be the result of structural damage and defects introduced during the grinding and separation processes. Also, in comparison with the virgin glass fibres which show no change in strength in relation with fibre length, there is a clear gradual decrease in the ultimate fibre strength with increasing fibre length (approximately 14% decrease in fibre strength between 5 and 15mm gauge length).

The results collected from these tests were modelled further using the well established Weibull distribution. The tensile strength of virgin and recyclate glass fibres in relation to their gauge length was analysed using the two-parameter Weibull model [12]. The probability of failure of a fibre of length L, for an applied strength σ, is given by the two parameter function:

$$P_f = 1 - \exp\left(-L\left(\frac{\sigma}{\sigma_0} \right)^m \right) \qquad (2)$$

Where σ_0 is a scaling parameter and represents the mean fibre strength and m is the shape parameter known also as Weibull modulus and represents the scatter of the fibre strength.

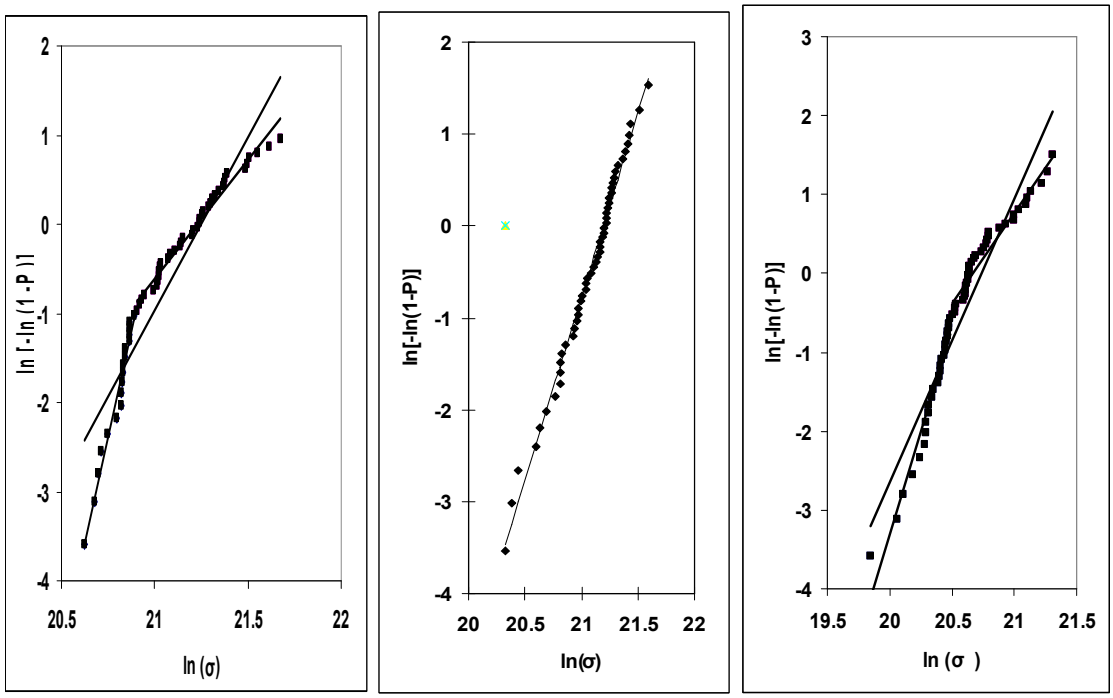


Figure 6: Two parameter Weibull analysis for virgin fibres at different gauge lengths: (a) 5mm; (b)10mm; (c) 15mm.

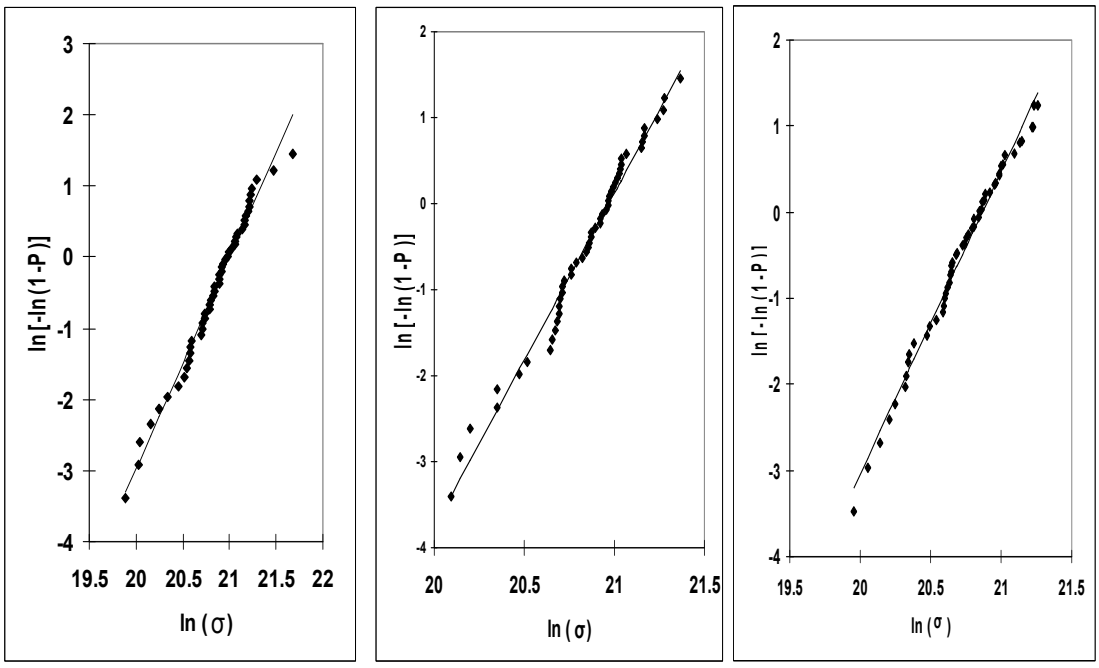


Figure 7: Two parameter Weibull analysis for recyclate fibres at different gauge lengths: (a) 5mm; (b)10mm; (c) 15mm.

The Weibull parameters as a function of fibre gauge length and type of fibres have been compiled in Table 2.

Gauge Length	Fibre	*m*	σ_0 *(GPa)*	R^2
5	Virgin	3.86	2.43	0.89
	Recyclate	3.16	1.92	0.98
10	Virgin	3.91	2.31	0.99
	Recyclate	3.88	1.84	0.98
15	Virgin	3.96	2.41	0.95
	Recyclate	3.51	1.66	0.99

Table 2: The value Weibull parameters for virgin and recyclate fibres at different gauge lengths.

In the case of virgin glass fibres, both parameters – scaling (σ_0) and shape (m) are less dependent of the fibre gauge length than the recyclate fibres, indicating a homogeneous material with uni-modal distribution of flaws. The decrease in value of the shape parameter (m) for recyclate fibres means that these fibres have a larger flaws density and a larger strength variance than the virgin fibres, less evenly distributed along the length of the fibres. A higher m value corresponds to a lower variance and when $m \rightarrow \infty$, the fibre variation approaches zero, for which the strength becomes independent of length. These results are not unexpected given the large amount of handling the recyclate fibres have been subjected to which could have lead to either surface flaws created by the uneven thick coating of SMC paste left on the fibres or defects within the fibre structure created by the grinding and separation processes. The importance of fibre surface defects has been recognized previously by other researchers [12, 13].

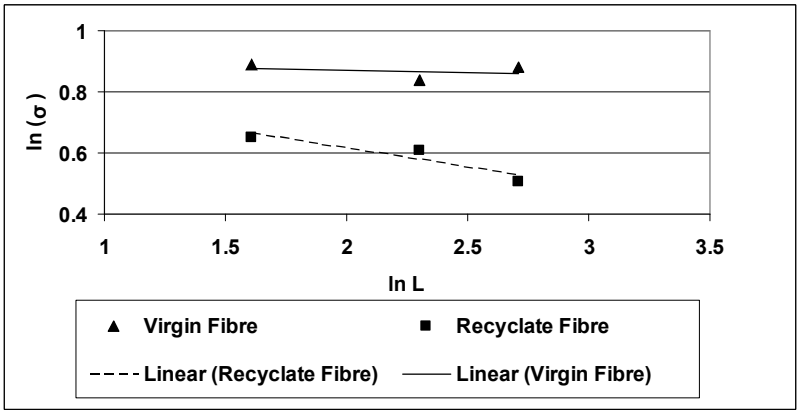
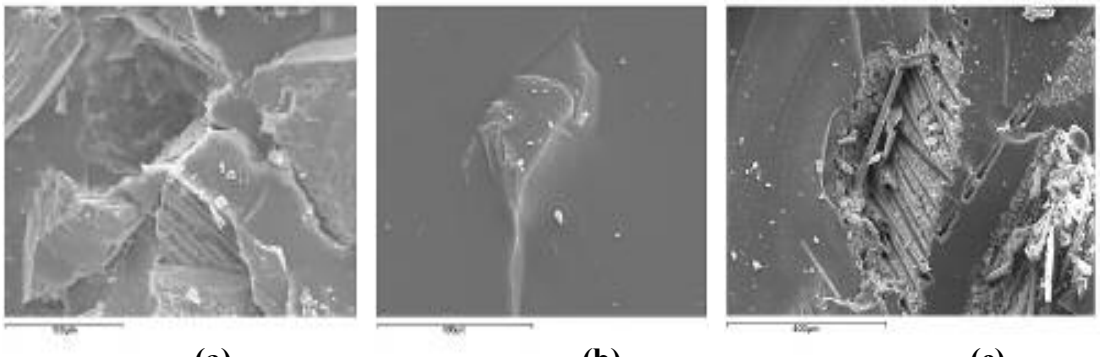


Figure 8: Mean tensile strength vs. gauge length for virgin and recyclate fibres.

4.3 Fracture surface analysis – SEM

SEM has been employed to investigate the bonding between a polyester resin matrix and filler particles of (i) cured polyester, (ii) $CaCO_3$ and (iii) recyclate particles. As shown in Figure 9a, the calcium carbonate reinforced matrix presents a range of fracture surfaces, failure cracks progress around particles and full debonding of particles has occurred indicating poor bonding. Conversely, in the case of 'self reinforced' polyester filled material (Figure 9b), the particles are much harder to detect where the crack propagates through the particles and there is no sign of de-bonding, indicating a stronger bond. This pattern is further illustrated in Figure 9c where the edges of the

recyclate particle appear to be well bonded to the new matrix, but it has failed internally.



(a) **(b)** **(c)**

Figure 9 Fracture surfaces of (a) $CaCO_3$ reinforced matrix, (b) self reinforced matrix, (c) recyclate reinforced matrix.

6. CONCLUSIONS

While the reuse of the cured resin and CaCO3 filler, components of ground recyclate should not affect mechanical properties, the weakening effect of reuse often reported is predominantly due to the condition of the recyclate fibres. Fibre damage during grinding results in a significant strength drop off, while the presence of residual debris on the recyclate fibre surfaces disrupts and diminishes fibre-matrix bond integrity on reuse.

The Weibull parameters revealed a hightened strength/length dependence in recyclate fibres compared with virgin fibres. This is probably a result of increased surface damage in recycled fibres, where a gauge length increase is accompanied by an increased likelihood of critical surface flaws being present in the region under load.

In spite of the excellent bonding observed between cured resin and virgin resin (in self reinforced structures), the SEM analysis revealed that when incorporating recyclate particles within virgin matrix, the particles fail within their structure and not at the matrix – particle interface.

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