CORRELATION OF MICRO- AND MACROCOMPOSITE PROPERTIES THROUGH FRAGMENTATION TESTING

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ABSTRACT: An advanced epoxy resin used in commercial applications has been pre-pregged with three different types of carbon fibres with differing surface finish. The interface dominant mechanical properties of the unidirectional materials have been assessed. Thus, interlaminar shear strength (ILSS), transverse strength and transverse modulus as well as unidirectional tensile strength and modulus have been measured. Single fibre fragmentation testing has been carried out using the same fibre and resin combinations. The correlation between fibre-matrix interface quality and the composite mechanical properties has been examined.

KEYWORDS: Single fibre fragmentation test, carbon fibre-reinforced epoxy, carbon fibre/epoxy composites, interlaminar shear strength, transverse strength.

INTRODUCTION

There is increasing interest in understanding the physical and chemical mechanisms responsible for fibre-matrix adhesion as well as its interrelation with the mechanical properties of advanced composites. Many earlier works on the development of composite materials considered fibre-matrix adhesion to be a necessary condition to ensure good composites performance. Some concentrated on increasing fibre-matrix adhesion through the use of surface treatments and sizings on fibres [1 - 4]. Several testing methods have been developed for measuring fibre-matrix adhesion using single fibres or bundles of fibres. The aim was to measure fibre-matrix adhesion and use it to predict the mechanical properties globally. There are three fundamental methods for measuring fibre-matrix adhesion, namely, single fibre fragmentation test, fibre pull-out test and fibre micro-indentation test. Among the three, the single fibre fragmentation test is commonly used because of the ease of sample preparation and the relevance of failure processes to real composites.

The interfacial shear strength is calculated from the fragmentation test by assuming that the interfacial shear stress is constant over the fragment length either due to either matrix yield in a completely plastic matrix or frictional stress transfer as a result of interfacial debonding. However, the results obtained with conventional data reduction techniques based on the constant

shear model of Kelly-Tyson [5] or the partial debonding model of Piggott [6] have several limitations. Different micromechanical features such as matrix yielding, transverse matrix cracking and interfacial debonding play an important role in the overall characterisation of the interface and are not properly accounted for in the existing data reduction techniques. This complexity in the fragmentation test makes correlation with the fibre surface chemistry and mechanical properties of the macrocomposites difficult [1, 7 - 9].

The object of this paper is to correlate the mechanical properties of real composites with their interfacial performance through single fibre fragmentation testing using Cumulative Stress Transfer Function (CSTF) technique proposed by Tripathi and Jones [10 - 12]. In this technique, the interfacial shear stress associated with the tensile stress in the individual fibre fragments are predicted from the plasticity effect model and the total tensile stress transferred to all of the fibre fragments at a particular matrix strain is calculated. Since, the CSTF technique is a direct measure of the efficiency of the stress transfer to the fibre across the interface, the correlation of the single fibre fragmentation (micromechanical) test with the macromechanical tests is considered to be easier.

EXPERIMENTAL PROCEDURE

The materials

HTA 5131, HTA 5001 and HTA 5000 are PAN based carbon fibres supplied by Tenax Carbon Fibres GmbH. HTA 5131 is surface treated and sized (TS), HTA 5001 is treated and unsized (TU) and HTA 5000 is untreated and unsized (UU).

MTM 60 epoxy resin from Advanced Composites Group Ltd. (ACG) was used for the single fibre fragmentation test and for the manufacturing of the pre-pregs. This is a medium temperature curing resin whose pre-pregs are mainly used for manufacturing structural components in motor sports. The resin was degassed in the vacuum oven before it was used in preparing the fragmentation test samples.

Single fibre fragmentation test

Single fibres were extracted at random from the carbon fibre tow, mounted on a steel wire, placed in a PTFE mould and embedded in a hot-cured MTM 60 resin. Care was taken to ensure that the fibre under test had not been touched and remained aligned during cure. The following cure cycle was used: 60°C for 30 minutes; 80°C to 120°C at 20°C per hour; 90 minutes at 120°C, followed by natural cooling to room temperature. The cured specimens were then ground and carefully polished. Test specimens were subjected to uniaxial tension at a displacement rate of 0.13 mm/min on a Mini Tensile Testing Machine (Micro Materials Ltd.). The fragmentation of single fibre was continuously monitored along its length. The test was stopped at each one percent of applied strain, starting from 4 % and all of the fragments in the specimen were photographically digitised. The fibre fragments that could not be accommodated on the video screen for digitisation (typically greater than 1.50 mm) were measured directly using a digital calliper fitted to the microscope. The test was stopped once the fragmentation had saturated.

Pre-preg manufacture

A laboratory drum winding machine was used to produce pre-preg from the three different carbon fibres. MTM 60 resin film with 50 gram per square metre (gsm) was used. The drum winding machine was set-up to obtain ~ 60 % fibre volume fraction.

Laminate manufacture

The pre-preg stack was vacuum bagged prior to curing in a press-clave. The cure cycle for the laminate in the press-clave is the same as cure cycle for preparing single fibre fragmentation test specimen.

Mechanical testing

All of the specimens for mechanical testing were cut from the laminate plate using a diamond-coated cutting wheel. Longitudinal tensile tests were carried out according to BS2782: Part 3: Method 320E: 1976, transverse tensile test according to ASTM D3039 (1976) and the short beam shear test according to ASTM D2344 (1984). All the tests were performed using the Mayes Universal Testing Machine model SM 200.

RESULTS AND DISCUSSION

Single fibre fragmentation test

The mechanical properties of the MTM 60 resin used for the fragmentation test are shown in Table 1. The other fibre properties used for the calculations of CSTF value are given in Table 2.

Table 1. Mechanical properties of the MTM 60 epoxy resin for the fragmentation test (standard deviation in brackets).

Tensile strength (MPa)	81.51 (2.34)
Tensile modulus (GPa)	3.20 (0.24)
Shear yield strength (MPa)	47.07
Failure strain (%)	7.83 (0.26)

Table 2. Fibre strength data for different carbon fibres used in the fragmentation test (standard deviation in brackets).

	TS	TU	UU
Tensile strength (GPa)	3.83 (0.78)	3.92 (0.98)	3.63 (0.93)
Elastic modulus (GPa)	189.17 (49.82)	192.16 (45.36)	185.06 (53.71)
Weibull modulus	5.10	4.35	4.12

Treated-sized fibre

The single fibre fragmentation test results for treated-sized fibre are shown in Table 3. During fragmentation of the treated-sized carbon fibres, it was observed that the first fibre fracture occurred at an applied strain greater than 2.6 %. There were very few fibre fragments shorter

than 1.50 mm at applied strain less than 3 %. The first detailed measurements of fibre fragment lengths were carried out at an applied strain of 4 %. Only a few fibres longer than 1.50 mm existed at 4 % strain. These fractured into smaller fragments as the applied strain is further increased. The broadest distribution in the fragment length from sample to sample was observed at an applied strain of 4 %. This narrowed as the applied strain was increased.

As the applied strain increased, the number of fibre fragments increased and the average fragment length decreased. Fragment saturation was recorded by comparing the number of fragments per unit length of the test specimen. We have assumed that a unit length of 10 mm for this purpose. It can be seen from Table 3 that the number of fragments per 10 mm increased with applied strain. From these results it can be concluded that saturation is not reached for treated-sized fibre. Furthermore, interfacial debonding was not observed during fragmentation so that the limitation in this case was the failure strain of the matrix.

Table 3. Fragmentation test results for the treated-sized carbon fibre in MTM 60 at different applied strains (standard deviation in brackets).

Strain (%)	4	5	6	7	8
Average fragment length (mm)	0.68 (1.13)	0.34 (0.17)	0.27 (0.08)	0.24 (0.07)	0.23 (0.06)
Average fragment aspect ratio	93.79	46.90	37.24	34.48	31.72
Critical fibre length (mm)	0.91	0.45	0.36	0.32	0.31
Average debonding length (mm)	0	0	0	0	0
Number of fragment per 10 mm	14.70	28.74	37.32	40.62	43.24
Apparent ISS (MPa)	22.36	51.23	67.49	77.70	81.76
CSTF value (MPa)	2974	2254	1871	1629	1533

Treated-unsized fibre

For the treated-unsized fibres the similar fragmentation profile was observed as shown in Table 4.

Table 4. Fragmentation test results for the treated-unsized carbon fibre in MTM 60 at different applied strains (standard deviation in brackets)

Strain (%)	4	5	6	7	8
Average fragment length (mm)	0.91 (1.03)	0.38 (0.14)	0.34 (0.09)	0.33 (0.09)	0.32 (0.09)
Average fragment aspect ratio	124	52.78	47.22	44.83	44.44
Critical fibre length (mm)	1.21	0.51	0.45	0.44	0.43
Average debonding length (mm)	0.004 (0.01)	0.02 (0.03)	0.03 (0.02)	0.04 (0.02)	0.07 (0.04)
Number of fragment per 10 mm	10.51	21.29	29.52	30.72	30.77
Apparent ISS (MPa)	16.95	49.63	56.90	59.03	61.31
CSTF value (MPa)	3863	2410	1796	1544	1374

However, in this case, the onset of the fragmentation began at higher applied strain, approximately at 3.0 %. Consequently fewer fragments were observed at an applied strain of 4 % than with the treated-sized fibres.As with any fragmentation experiment, the fragment length distribution became narrow as the applied strain was increased. It can be seen in Table 4 that the

number of fragments per 10 mm reached a plateau at 6 % strain of which was indicative of saturation. In this case, interfacial debonding of the fragments could be observed during fragmentation at strains close to onset fibre fracture.

It can be seen in Table 3 and 4 that the average fragment length for the treated-sized fibres was less than that of treated-unsized fibres. The limitation to fragmentation of the treated-unsized fibres was clearly the onset of debonding. Therefore, sizing of the fibres appears to have modified the interfacial bond.

Untreated-unsized fibre

The single fibre fragmentation results for untreated-unsized fibre are shown in Table 5. In this case, saturation was achieved between an applied strain of 5-6 %. Extensive interfacial debonding was observed for these fibres and was already significant at a strain of 4 %. This is to be expected since the untreated-unsized fibres should have the lowest fibre-matrix adhesion.

Table 5. Fragmentation test results for the untreated-unsized carbon fibre in MTM 60 at different applied strains (standard deviation in brackets).

Strain (%)	4	5	6	7	8
Average fragment length (mm)	0.85 (0.95)	0.37 (0.14)	0.34 (0.10)	0.34 (0.10)	0.34 (0.10)
Average fragment aspect ratio	114.40	49.80	45.76	45.76	45.76
Critical fibre length (mm)	1.13	0.49	0.45	0.45	0.45
Average debonding length (mm)	0.012 (0.02)	0.030 (0.04)	0.05 (0.04)	0.07 (0.06)	0.09 (0.07)
Number of fragment per 10 mm	12.41	26.95	27.50	27.56	27.56
Apparent ISS(MPa)	18.01	50.63	56.24	56.24	56.24
CSTF value (MPa)	3588	2101	1659	1383	1200

Interfacial shear strength and cumulative stress transfer function (CSTF)

A comparison of the Kelly-Tyson ‘interfacial shear strength’ for the treated-sized, treated-unsized and untreated-unsized fibres at similar applied strains shows that treated-sized fibre has higher interfacial shear strength compared to treated-unsized and untreated-unsized fibres as shown in Figure 1.

The predictions of interfacial quality by the Kelly-Tyson model are consistent with the experimental observations on the extent of fibre-matrix debonding. Furthermore, the treated-sized fibres exhibited the shortest average fragment length and did not reach saturation prior to failure. It can be seen that the estimate of ‘interfacial shear strength’ is much higher than the shear yield strength of the matrix, which was calculated to be 47.07 MPa from von Mises criterion. For the untreated-unsized fibres, interfacial debonding was observed during fragmentation so that the ‘interfacial shear strength’ should be lower than the shear yield strength of the matrix. But this is not the case. However, complete debonding was not observed. For neither system is the criterion of a constant shear stress achieved so that the Kelly-Tyson analysis is invalid. This has been highlighted by several workers [10, 12 - 15].

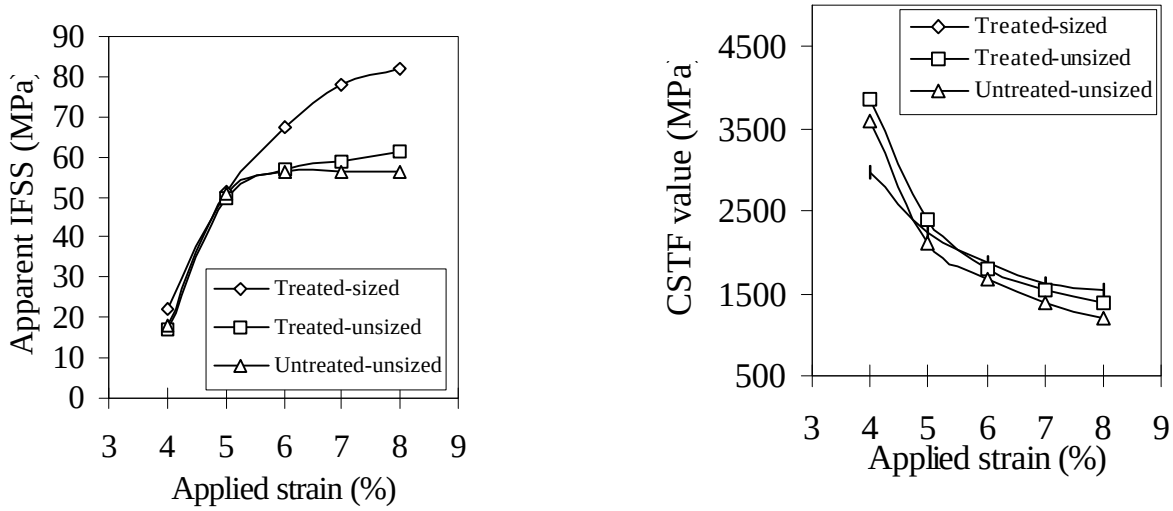

Fig. 1. Interfacial shear strength and CSTF values for the three different fibres in MTM 60.

Meanwhile, the comparison of CSTF values shows that treated-sized fibre has the highest CSTF value over applied strain of 6 % to 8 %. The reduction in CSTF value with applied strain for all the fibres is a consequence of the assumption that more stress is transferred to a longer fragment. Therefore, the treated-unsized fibre which has a longer average fragment length especially over the strain range of 4 and 5 % has the larger value of CSTF. The value of CSTF is also dependent on the extent of debonding and matrix yielding during fragmentation. Interfacial debonding reduces the value of CSTF significantly as shown by the trends for treated-unsized and untreated-unsized fibres.

The most interesting observation is the fact that the data for the treated-sized fibres are below the other lines at low strain but above them at high strain. This is indicative of a differing mechanism of stress transfer. In this case debonding was not observed and yielding at the interphase is clearly responsible. The sizing resin has therefore modified the resin matrix near the fibre interface. We have showed elsewhere[16] that yielding leads to a delayed build up of stress in the fibre leading to less fragmentation. Once debonding sets-in for the untreated-unsized fibres this has a much larger effect on the stress transfer function, and the continued fragmentation at high strains is less efficient.

The CSTF technique is clearly more sensitive to subtle changes in interfacial quality and can be seen to be more appropriate than the constant shear approach to the quantification of the interfacial response. It also demonstrates that it is easier to differentiate between interface deformation mechanisms, using the CSTF concept.

Mechanical properties of unidirectional laminates

The mechanical properties of the three different composites are shown in Table 6.

Table 6. Mechanical properties of MTM 60/carbon fibre composites (standard deviation in brackets).

Fibre treatment	TS	TU	UU

Fibre volume fraction (V_f)	67.2	64.0	62.5
Tensile strength (MPa)	1615 (80)	1496 (48)	1249 (69)
Tensile modulus (GPa)	163.0 (9.2)	158.3 (13.1)	143.2 (7.5)
Transverse tensile strength (MPa)	52.5 (4)	39.3 (5.7)	33.50 (2)
Transverse modulus (GPa)	9.53 (0.82)	9.28 (1.0)	9.62 (0.44)
Interlaminar shear strength (MPa)	66.3 (2.5)	63.1 (3.2)	55.0 (1.4)

For comparison purposes, all of the mechanical properties shown in Table 6 have been linearly scaled to the average fibre volume fraction of all the composites of 64.6 % in Figure 2, 3 and 4.

A comparison of the longitudinal moduli of the three composites shows that there is no significant difference (Fig. 2). This shows that modulus, which is a fibre dominated property is insensitive to the differences in the quality of fibre-matrix adhesion

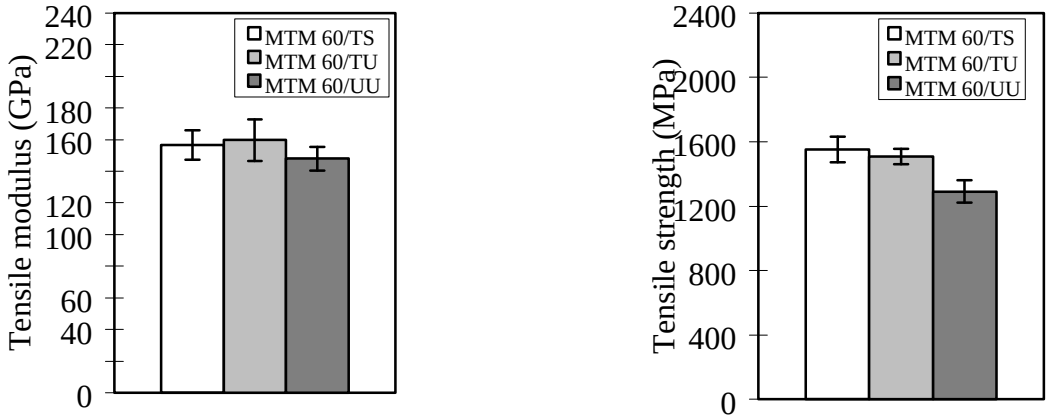

Fig. 2. Longitudinal tensile modulus and tensile strength of MTM 60/TS, MTM 60/TU and MTM 60/UU composites.

significantly different but MTM 60/UU composite has a lower tensile strength. The failure modes of the three system were found to be different. The MTM 60/TS composite failed with

leading to brittle fracture. similar observations were reported elsewhere [7, 8] for composites with good fibre-matrix adhesion. Meanwhile, the failure of MTM 60/TU composite showed

for fibre with intermediate fibre-matrix adhesion was reported by several workers [7, 8, 17]. The failure of MTM 60/UU, on the other hand, showed more internal damage rather than

transfer the load so efficiently and the stored strain energy is not large enough to cause extensive fibre-matrix splitting during the fracture. This explains why MTM 60/UU composite

With transverse tensile strength, failure is dominated by fracture of the matrix and/or interface, the influence of the interface is very clear. MTM 60/TS composite had the highest transverse

since in a transverse tensile test, the interfacial bond between fibre and matrix has a direct

influence on the ease of fracture. Considering that MTM 60/TU and MTM 60/UU composites have a tendency to interfacial debonding at fibre break as shown in results of the fragmentation test , we could expect lower transverse tensile strengths than the MTM 60/TS composite. As with longitudinal tensile modulus, the transverse tensile modulus was found to be insensitive to the fibre-matrix adhesion. The results for transverse tensile strength and modulus shown in Fig. 3 are in agreement with those reported earlier by several workers [7, 9].

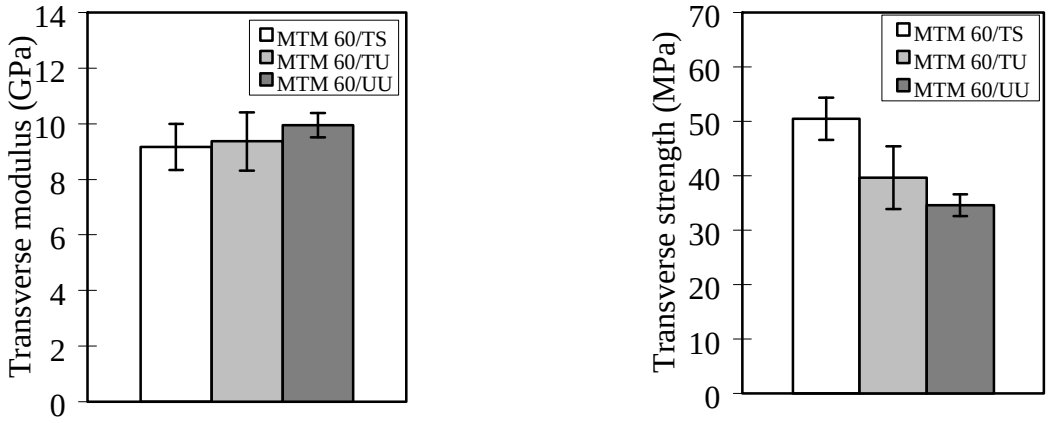

Fig. 3. Transverse modulus and transverse tensile strength of MTM 60/TS, MTM 60/TU and MTM 60/UU composites.

Interlaminar shear strengths (ILSS) obtained from the short beam shear tests are shown in Fig. 4. It can be seen that the interlaminar shear strength is not sensitive enough to show significant change in fibre-matrix adhesion for MTM 60/TS and MTM 60/TU composites but does shows significantly lower ILSS for MTM 60/UU composite. The results for MTM 60/TS and MTM 60/TU composites are not entirely unexpected. It was reported earlier that interlaminar shear strength is insensitive to fibre-matrix adhesion [9]. Another worker [7] also found only a slight improvement in interlaminar shear strength from treated-unsized to treated-sized composites.

It is demonstrated that there was a close correlation between the interface dominated macrocomposite properties and the CSTF value. The Kelly-Tyson data analysis did not differentiate between the changes in interface quality that affected the failure processes in the macrocomposite.

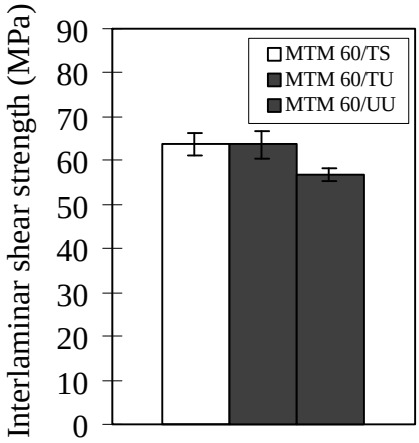

Fig. 4. Interlaminar shear strength of MTM 60/TS, MTM 60/TU and MTM 60/UU composites.

CONCLUSIONS

This study has shown that single fibre fragmentation can be carried out in a matrix resin used for the preparation of laminates. The Cumulative Stress Transfer Function (CSTF) can be used successfully to quantify fibre-matrix adhesion from the data of single fibre fragmentation test. It proved unnecessary to reach saturation in the fragmentation process.

A good correlation between micro-and macromechanical properties was demonstrated.

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REFERENCES

1. Cheng, T. H., Jones, F. R. and Wang, D. ''Effect of Fibre Conditioning on the Interfacial Shear Strength of Glass-Fibre Composites''. *Composites Science and Technology*, Vol. 48, 1993, pp. 89-96.

2. Cheng, T. H., Zhang, J., Yumitori, S, Jones, F. R. and Anderson, C. W. ''Sizing Resin Structure and Interphase Formation in Carbon Fibre Composites''. *Composites*, Vol. 25, Number 7, 1994, pp. 661-670.

3. Yumitori, S, Wang, D. and Jones, F. R. ''The Role of Sizing Resins in Carbon Fibre-Reinforced polyethersulfone (PES)''. *Composites*, Vol. 25, Number 7, 1994, pp. 698-705.

4. Cheng, T. H. ''The Role of Sizing Resin on the Micromechanics of Fibre Composites''. *Ph.D. thesis*, 1994, The University of Sheffield, U.K.

5. Kelly, A. and Tyson, W. R., "Tensile Properties of Fibre Reinforced Metals: Copper/Tungsten and Copper/Molybdenum", *Journal of Mechanics and Physics of Solids*, Vol. 13, 1965, pp. 329-350.

6. Piggott, M. R., "Expressions Governing Stress-Strain Curves in Short Fibre Reinforced Polymers", *Journal of Materials Science*, Vol. 13, 1978, pp. 1709-1716.

7. Drzal, L.T. and Madhukar, M., "Fibre-Matrix Adhesion and Its Relationship to Composite Mechanical Properties", *Journal of Materials Science*, Vol. 28, 1993, pp. 569-610.

8. Ivens, J., Wevers, M. and Verpoest, I., "Influence of Carbon Fibre Surface Treatment on Composite UD Strength", *Composites*, Vol. 25, No. 7, 1994, pp. 722-728.

9. Hoecker, F. and Karger-Kocsis, J., "Effect of the Interfaces on the Mechanical Responce of CF/EP Microcomposites and Macrocomposites", *Composites*, Vol. 25, No. 7, 1994, pp. 729-738.

10. Tripathi, D., Chen, F. and Jones, F. R., "The Effect of Matrix Plasticity on the Stress Fields

in a Single Filament Composite and the Value of Interfacial Shear Strength Obtained from the Fragmentation Test", *Proceeding of the Royal Society A: Mathematical and Physical Sciences,* Vol. 452, 1996, pp. 621-653.

11. Tripathi, D., "Micromechanics of Interfaces in Short Fibre Reinforced Polymer Composites", *Ph.D. Thesis, The University of Sheffield*, 1995.

12.Tripathi, D. and Jones, F. R., "Measurement of Load Bearing Capability of the Fibre/Matrix Interface by Single Fibre Fragmentation", *Composites Science and Technology*, Vol. 57, 1997, pp. 925-935.

13. Melanitis, N, Galiotis, C., Tetlow, P. L. and Davies, C. K. L., "Interfacial Stress Distribution in Model Composites Part 2: Fragmentation Studies on Carbon Fibre Epoxy System", *Journal of Composite Materials*, Vol 26, 1992, pp. 574-610.

14. Feillard, P., Desarmot, G. and Favre, J. P., ''A Critical Assessment of the Fragmentation Test for Glass/epoxy System'', *Composites Science and Technology*. Vol. 49, 1993, pp. 109-119.

15. Feillard, P., Desarmot, G. and Favre, J. P., "Theoretical Aspects of the Fragmentation Test", *Composites Science and Technology*, Vol. 50, 1994, pp. 265-279.

16. Lane, R., Hayes, S.A. and Jones, F. R., ''Modelling of the Efficiency of Strain Transfer Across an Interphase Region in Fibre Reinforced Composites'', *Composite Interfaces*, in press.

17. Bader, M. G., ''Tensile Strength of Uniaxial Composites'', *Science and Engineering of Composite Materials*, Vol. 1, 1988, pp. 1 - 11.