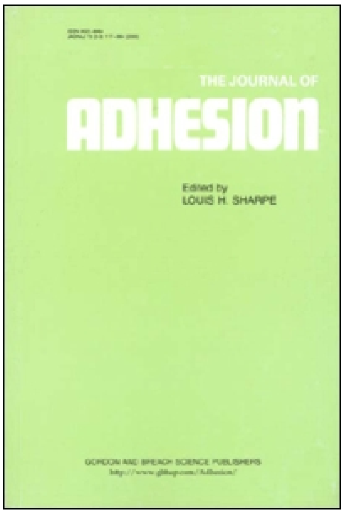

The Journal of Adhesion
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713453635

The Effect of Fibre Surface Treatment on the Failure of Continuous Carbon Fibre/Epoxy Resin Composites

K. L. Pickering[a]; M. G. Bader[a]
[a] Department of Materials Science and Engineering, University of Surrey, Guildford, Surrey, UK

To cite this Article Pickering, K. L. and Bader, M. G.(1994) 'The Effect of Fibre Surface Treatment on the Failure of Continuous Carbon Fibre/Epoxy Resin Composites', The Journal of Adhesion, 45: 1, 161 — 172
To link to this Article: DOI: 10.1080/00218469408026636
URL: http://dx.doi.org/10.1080/00218469408026636

J. Adhesion, 1994, Vol. 45, pp. 161–172
Reprints available directly from the publisher
Photocopying permitted by license only

Printed in the United States of America

The Effect of Fibre Surface Treatment on the Failure of Continuous Carbon Fibre/Epoxy Resin Composites*

K. L. PICKERING and M. G. BADER**

Department of Materials Science and Engineering, University of Surrey, Guildford, Surrey GU2 5XH, UK

(Received December 28, 1992; in final form April 7, 1993)

The failure mechanisms of a composite, consisting of continuous, aligned, high strength, polyacrylonitrile (PAN) based carbon fibre in an epoxy resin, under uniaxial tension, have been studied. In order to study the effect of the interphase/interface strength, six different levels of an electrochemical fibre surface treatment were used. Single tows containing approximately 12,000 treated carbon fibres were impregnated to produce composite rods with a fibre volume fraction of 0.55. Lengths of this impregnated tow were also set in the centre of glass-fibre/epoxy resin composite coupons which were used to study the mechanisms of failure of the embedded tows. Acoustic emission was used to monitor all samples and bundle failure was found to occur after a build-up of sub-critical damage events as previously modelled.[1] Microdebond tests demonstrated an initial increase of interfacial strength which levelled out at the higher levels. In impregnated samples with high surface treatments, catastrophic failure occurred with the crack propagating approximately perpendicular to the fibre direction. However, in samples with lower fibre surface treatments, longitudinal splitting (not accounted for in current models), occurred, meaning that a greater length of composite was involved in the final failure process. Acoustic emission has been shown to have an approximately direct relation with the predicted number of single fibre breaks in composite test-pieces; however, there was no significant difference attributable to the different surface treatments. The hybrid test coupons allow a detailed assessment of the failure mechanisms within the impregnated carbon tow. The failure strains of the embedded tow is some 5% higher than that of unsupported tow. The Weibull modulus is of the same order.

KEY WORDS carbon fibre; tensile strength; uniaxial composites; failure mechanisms; surface treatment; acoustic emission.

INTRODUCTION

Failure of continuous-fibre composites is generally considered to be a consequence of an accumulation of single-fibre failures occurring in a sporadic sequence. These are generated as the applied strain exceeds the local fibre failure strain causing failure at the weakest points, leading to a redistribution of stress around each fibre break. Stress is transferred back into the broken fibre away from these fibre-breaks

*Presented at the International Symposium on "The Interphase" at the Sixteenth Annual Meeting of The Adhesion Society, Inc., Williamsburg, Virginia, U.S.A., February 21–26, 1993.
**Corresponding author.

by shear stresses in the resin transferred across the interphase/interface. There are also stress concentrations on neighbouring surviving fibres, increasing their probability of failure. Failure of the composite is initiated when a critical number of neighbouring fibre breaks have occurred, this configuration generally termed a "critical i-plet". The distance along the fibre length over which stress builds up from a fibre break, commonly referred to as the transfer length, will affect the length of the neighbouring fibres affected by the break, and the degree to which they are overstressed. Increasing the strength of the interphase/interface will reduce the transfer length, so that a shorter length of fibre is affected by the neighbouring break, however, there will be a greater stress concentration over this shorter length. Thus, although the shorter overload segment will be stronger, this may be balanced by the effect of the higher stress concentration. This paper reports on experimental work designed to study the effects of varying the interphase/interface strength.

EXPERIMENTAL

High strength PAN-based carbon fibre [Tenax HTA], supplied by Akzo Faser AG was used for this investigation. The fibres were of 7 μm nominal diameter in tows of 12,000 filaments, all originating from the same batch of precursor. As received the fibres had been subjected to various levels of an electrochemical surface treatment, designated as 0, 0.25, 0.5, 0.75, 1.0, and 2.0, where these values are related to the applied current and 1.0 is representative of the manufacturer's standard surface treatment.

The main impregnating resin was based on a formulation of a tetra-functional epoxy [MY720] with diphenyl-diamino-sulphone [DDS] curing agent and a boron tri-fluoride-based catalyst supplied by Ciba-Geigy Ltd.

Single fibres of each surface treatment were tensile tested using a novel testing machine developed at the University of Surrey.[2] About 25 measurements were made on fibres from each batch, all at a gauge length of 62 mm. The data were analysed according to the Weibull distribution.

The strength of the fibre/resin interface was measured by means of a microdebond test.[3] The debonding stress increased from 18 MPa at 0 surface treatment, and increased to 67 MPa at the 0.75 level of surface treatment. No further increase of debond stress was observed at the 1.0 and 2.0 surface treatment levels.

Surface treatment of the fibre has also been carried out.[4] X-ray Photoelectron Spectroscopy (XPS) showed a constant increase in the oxygen:carbon ratio with increased surface treatment up to the highest level.

Impregnated tow was continuously produced using an apparatus designed inhouse. Here the tow of 12,000 fibres was hauled by means of motor-driven rollers, from its spool, through a heated resin bath, and then through a heated die, to improve consolidation and to control the fibre volume fraction. The impregnated tow was then pulled through a tube oven where it was cured beyond its gel-point, after which it could be cut to any required length. The resulting material is a cylindrical rod of composite with a diameter of approximately 1 mm and a fibre volume

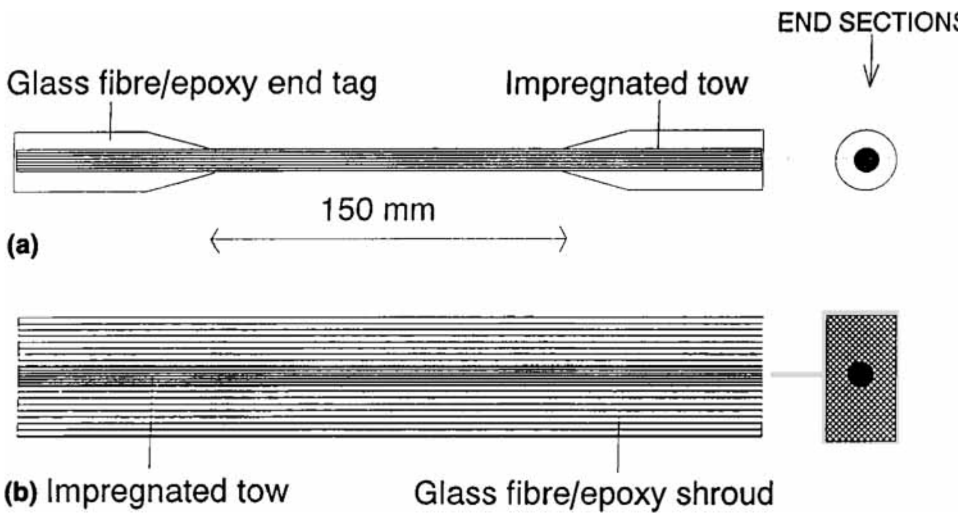

FIGURE 1 Sample Schematic Showing: a) End-tagged impregnated bundle; b) Hybrid.

fraction of 0.55. These lengths of composite were then prepared for tensile testing by fitting with braided glass-fibre/epoxy end tags and finally cured. The final cure schedule was 1 hour at 135°C followed by 2 hours at 170°C. This ensured the integrity of the end tags. The gauge length was 150 mm (see Fig. 1a).

These impregnated bundles were tensile tested on a standard Instron 4501 machine. About 30 samples at each surface treatment were tested. Acoustic emission was monitored during tensile testing of some test pieces in order to obtain information about the accumulation of damage leading to composite failure (see Fig. 2). The tensile strength was calculated on the basis of the nominal total fibre cross-section in the rod. The data were again analysed on the basis of a Weibull distribution.

Hybrid samples were also produced; impregnated tows, after production as described above, were fully cured as described above, and then set into parallel-sided coupons of glass fibre in an anhydride-cured, DGEBA [MY750]-based resin so that they were surrounded along the length of the tow by continuous glass fibre in epoxy resin (see Fig. 1b). This system was chosen because it has a higher strain to failure than the impregnated carbon tow, so that when the impregnated tow fails it remains supported by the surrounding GRP material. This preserves the failure zone for subsequent microscopic study and also has the advantage of absorbing some of the energy released at failure which limits the consequential damage. Due to the transparency of the glass fibre/MY750 resin, failures on the impregnated tow can be observed visually. A leaky mould technique[5] was used to achieve this configuration (see Fig. 3). Glass fibre was laid into an open-ended mould with a length of impregnated tow at its centre. Resin was then poured into the mould and the top half of the mould was placed in position. The whole mould was then placed into a hot press where tension was applied to the glass fibres and impregnated tow by means of weights attached to, and hung from, their ends to maintain their

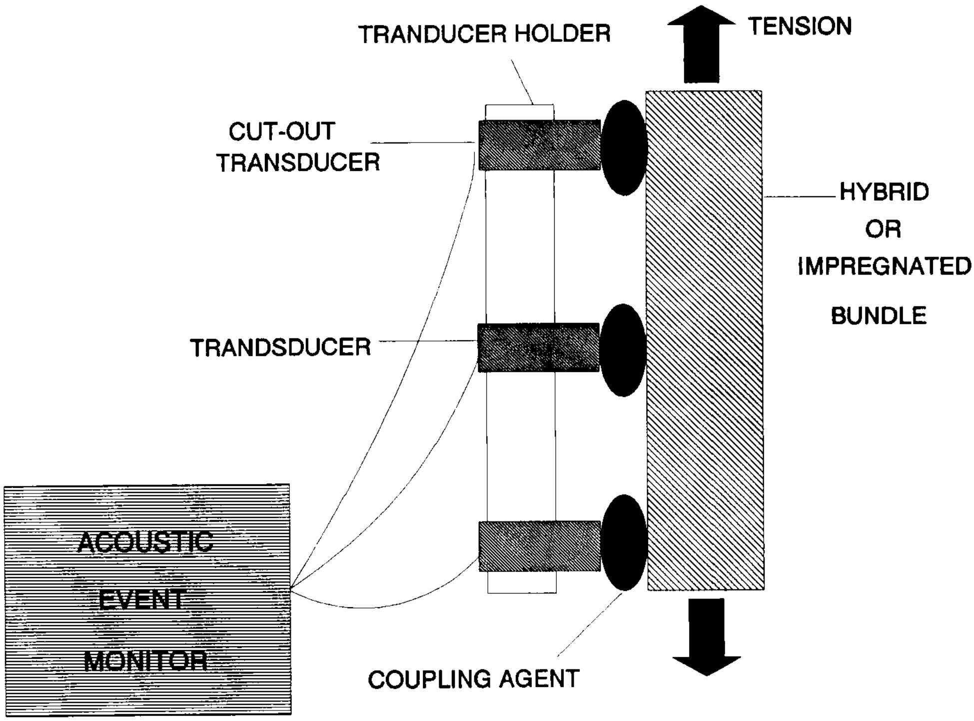

FIGURE 2 Acoustic monitoring schematic.

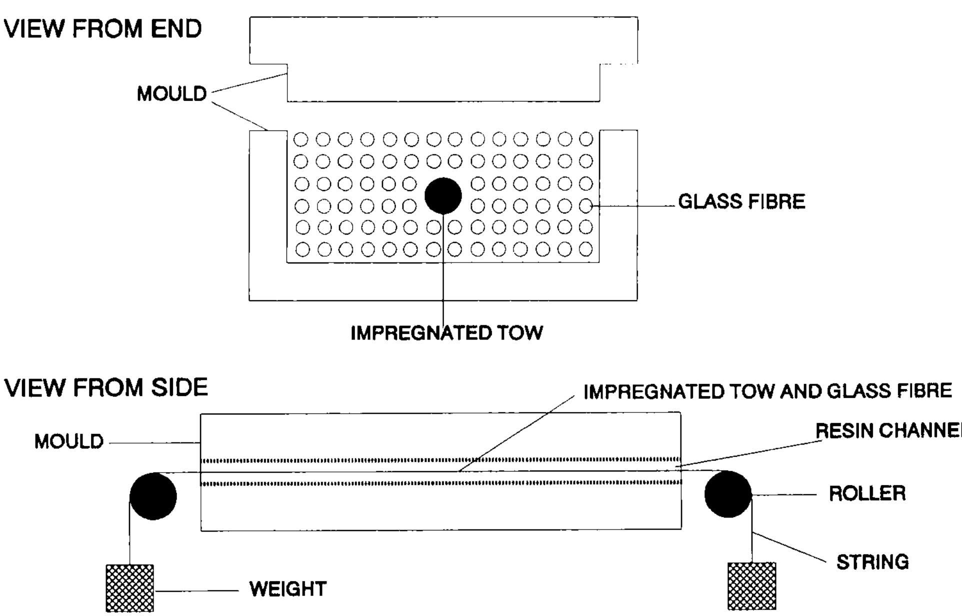

FIGURE 3 Hybrid production schematic.

alignment. The weight on the impregnated tow was 1 kg, which translates to a stress of 23 MN/m^2 applied to the carbon fibre which would result in a strain of less than 0.1%. The mould was then heated to lower the viscosity of the resin and the top half of the mould was then closed onto shims to give the correct thickness. The consolidated moulding was then cured in the hot press. The cure cycle was 3 hours at 120°C followed by 3 hours at 150°C.

Tensile testing of hybrids was carried out using an Instron 1195 testing machine. During testing the coupon was illuminated from behind using a halogen lamp. A polariser was placed between the light source and the coupon, and an analyzer between the coupon and the operator. Under conditions of crossed polarisers, the strain birefringence at a tow break rendered it easily visible. As the coupon was extended in tension, the strain, and position of each tow-break within a defined gauge length was recorded. Initially, the test pieces were extended until the first fracture of the impregnated tow occurred. However, some samples have now been extended up to a strain of 2%, when up to 6 individual tow failures occurred along the gauge length. All samples were monitored using acoustic emission during tensile testing. As yet, only a small number of samples at each surface treatment level have been tensile tested.

RESULTS AND DISCUSSION

For the single fibre tensile tests the force/extension curves show an increase in Young's modulus with increase in strain. This well-known phenomenon is considered to be due to structural realignment within the fibre. The secant modulus over the strain range was found to be between 196 and 223 GPa. Only small changes (3.24 to 3.48 GPa) in the characteristic strength of the single fibres was observed with the different surface treatments and no obvious pattern emerged. The average Weibull modulus was 5.3, again with no apparent trend due to surface treatment.

For the impregnated bundles (see Table I) there is a small decrease in strength with increased surface treatment, with the exception of composites with the highest treatment (2.0) which had a higher strength than those treated to the 1.0 and 0.75

TABLE I
Impregnated and hybrid tensile test data.
Data for impregnated bundles of gauge length 150 mm including average strength weak-link scaled for a gauge length of 180 mm

Surface treatment	Normalised average strength (GPa)	Weibull modulus	Effective strength for 180 mm
0	3.88	26	3.85
0.25	3.80	24	3.77
0.5	3.81	34	3.79
0.75	3.75	30	3.73
1.0	3.66	25	3.63
2.0	3.76	30	3.74

levels. The reasons for this anomaly are not understood. Weibull moduli ranged from 24 to 34; these high values give confidence in the data. However, the strength variations are quite small and not significant in a strict statistical analysis. The decrease of strength with increased surface treatment can be explained by the increase in interfacial shear strength that occurs with increase in surface treatment. This localises the stress around a fibre failure and increases the probability of adjacent fibre failure, resulting in a lower tensile strength.

The mode of failure of the impregnated bundles was seen to change dramatically with treatment level. At the highest levels of treatment the failure path runs approximately perpendicular to the fibre direction. It would appear that the stronger interface allows the crack to propagate with greater ease from fibre to matrix and into the neighbouring fibre. At the low levels of surface treatment, splitting was a prominent feature of failure (see Fig. 4). This is attributed to the relative ease with which the crack may be diverted along the relatively weaker interface. Acoustic events were observed to build up exponentially to failure (see Fig. 5). It is assumed, at this stage, that the principal cause of the acoustic emission is from fibres breaking. The number of expected fibre breaks was calculated by using the Weibull equation and the data from the single fibre experiments. This is shown in Figure 5 to give a very close fit to the observed acoustic event data. Although the one-to-one correlation may be considered fortuitous due to the arbitrary choices of amplification and threshold level for the acoustic signal, the fact that there is a proportional correspondence supports the proposition that the fibres within the composite have a strength distribution similar to the single fibres tested in free air. Examining the data of all

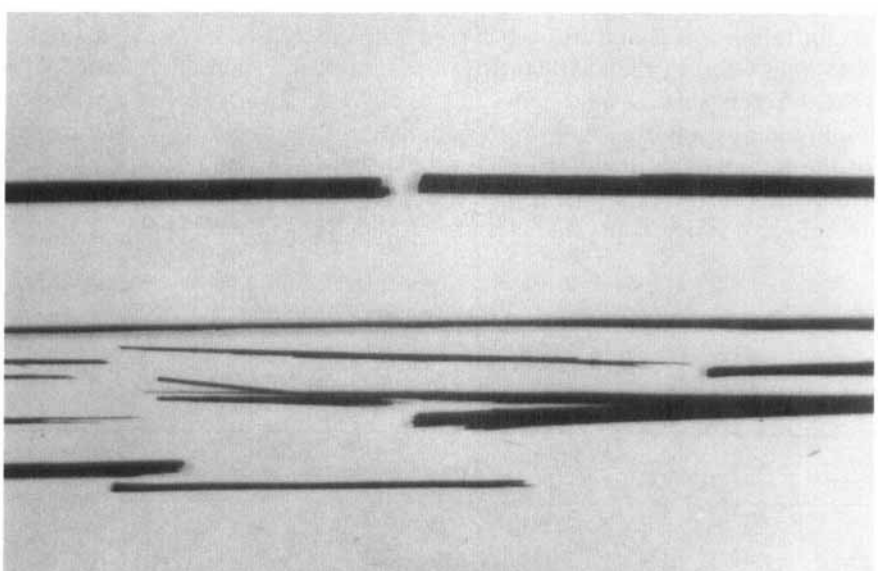

FIGURE 4 Tensile tested impregnated bundles showing the effect of surface treatment. The upper sample is of relatively high surface treatment (0.75) and the lower sample is of untreated fibre.

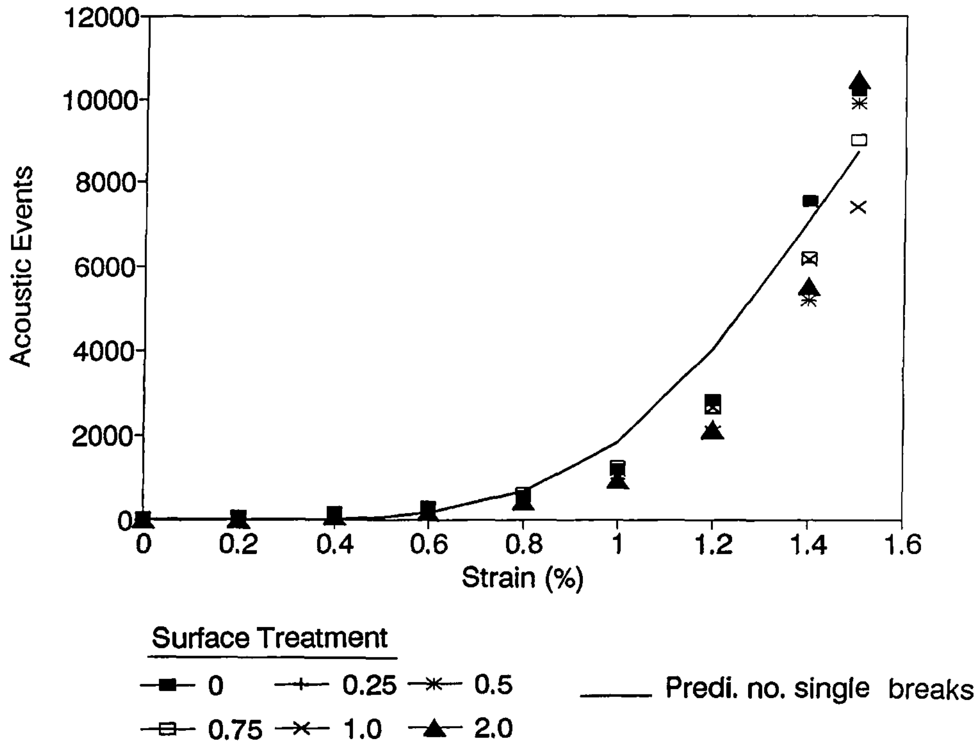

FIGURE 5 Accumulated acoustic events *versus* strain for impregnated bundles.

surface treatments, there was a general trend of increasing failure strain with an increase in accumulated acoustic events (see Fig. 6), implying that samples showing higher failure strains sustained more single fibre damage, but that the critical damage event was delayed to the higher strain level. Acoustic emission, therefore, gives no more indication than strain as to when failure is about to occur.

The first failure of impregnated tows in the hybrid samples showed a trend similar to that of the impregnated bundles, in that there was a small general decrease in failure strain as surface treatment level increased with the exclusion, again, of the highest treatment level (see Table II). These first failures were found to occur consistently at failure strains of about 1.07 times as high as those for impregnated tows tested in air. This is consistent with the well-established "hybrid effect" studied by many workers.[6,7] Due to the small amount of data obtained at this time, the data for all surface treatments have been compiled in one Weibull plot (see Fig. 7).The fit shows a reasonable straight line plot giving the Weibull parameter of 31, this being within the range for the impregnated bundle values. At failure, a zone of debonding was observed to form between the carbon bundle and the surrounding glass/epoxy (see Fig. 8). The acoustic emission was observed to build up sharply to first failure as with the impregnated bundles, again indicating an increasing number of single fibre breaks. The mode of failure observed showed similarities to that for

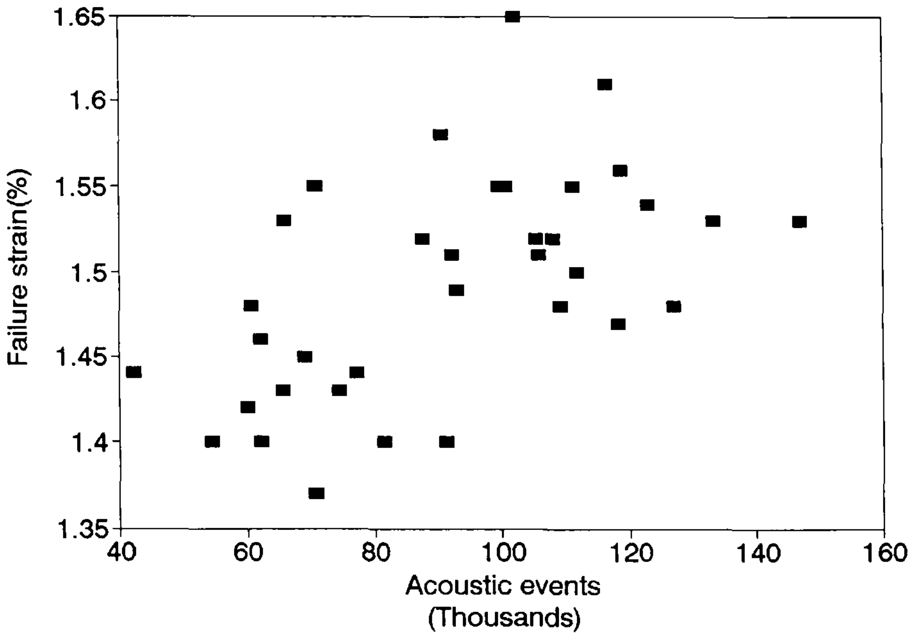

FIGURE 6 Failure strain *versus* total accumulated events for impregnated bundle samples.

TABLE II
Impregnated and hybrid tensile test data.
Data for hybrids showing relation to impregnated bundles

Surface treatment	Average strain at 1st failure ϵm (%)	Average stress in 1st failure σm (GPa)	σm (hydrid 180 mm) / σm (imp. bundle 180 mm)
0	1.8	4.25	1.10
0.25	1.66	3.97	1.05
0.5	1.74	4.11	1.08
0.75	1.66	3.97	1.06
1.0	1.63	3.83	1.06
2.0	1.70	4.00	1.07
			1.07

the impregnated tows. The longitudinal splitting was found to be greatest at low surface treatment levels, giving a longitudinally-extended major crack. Failure at higher treatment levels was generally characterised by the major crack running almost perpendicular to the fibre direction (see Fig. 9). Other fibre breaks, which did not participate in the tow failure, were also visible but with no significant separation between their ends. Multiple neighbouring fibre breaks were generally observed to occur where fibres had very little spacing between them (see Fig. 10).

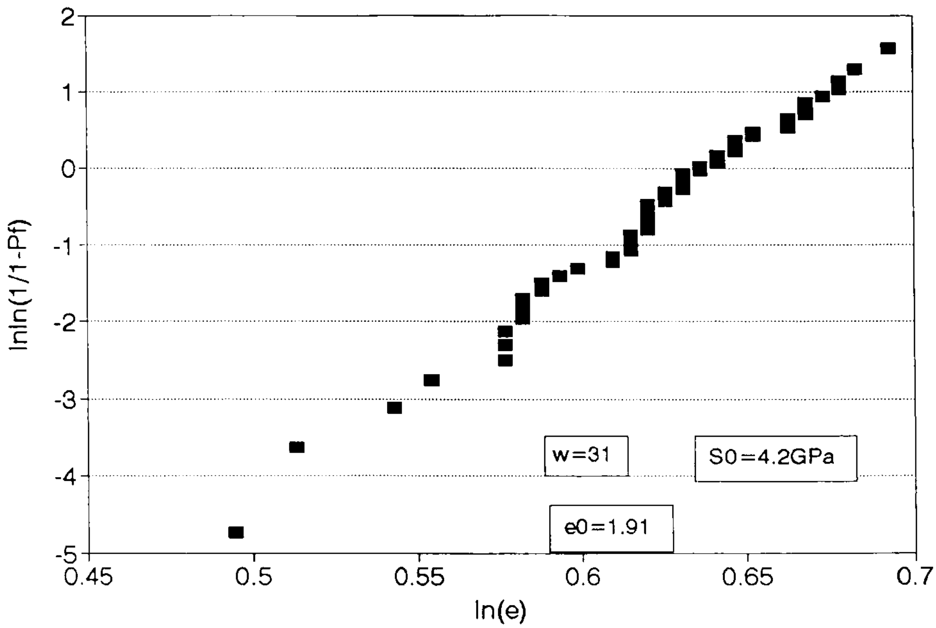

FIGURE 7 Weibull plot for hybrids showing all surface treatments.

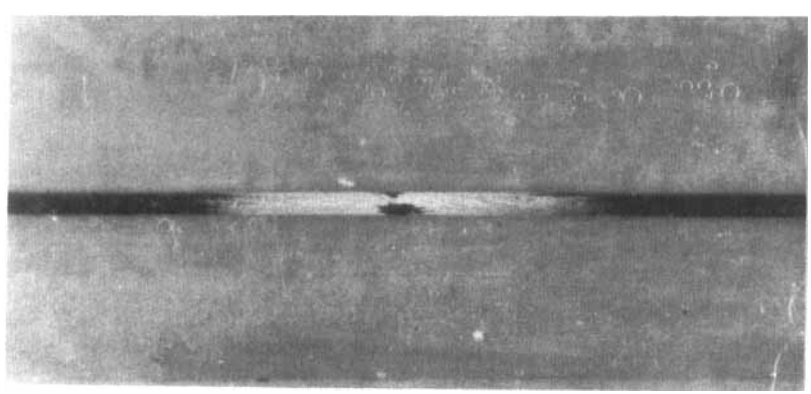

FIGURE 8 Debonding of impregnated tow from glass reinforced plastic.

CONCLUSIONS

The tensile strength of the impregnated bundle composites was generally found to decrease with surface treatment. This must be assumed to be due mainly to the increased interphase/interface strength rather than to a reduction of the fibre strength, since no obvious trend with respect to surface treatment was observed

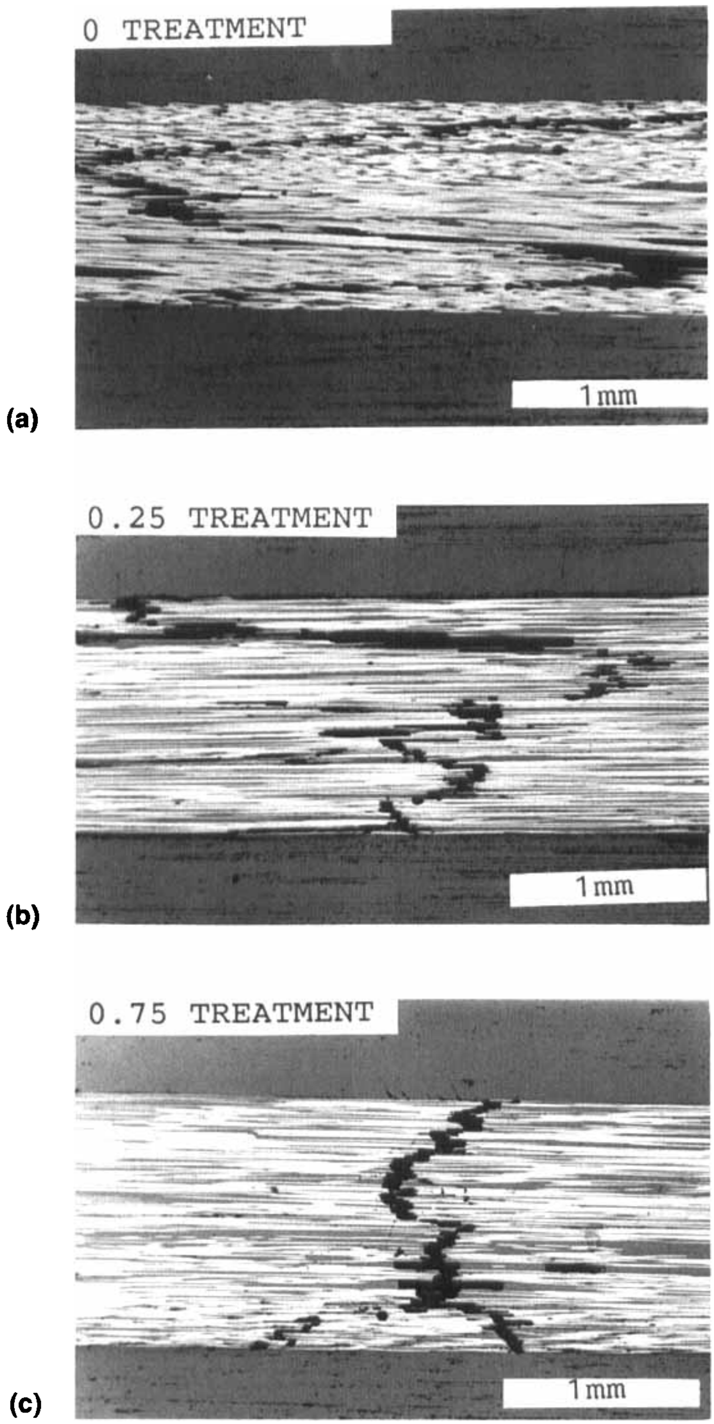

FIGURE 9 Hybrid samples showing characteristic failure modes at: a) Low (0), b) Medium (0.25) and c) High (0.75) surface treatments.

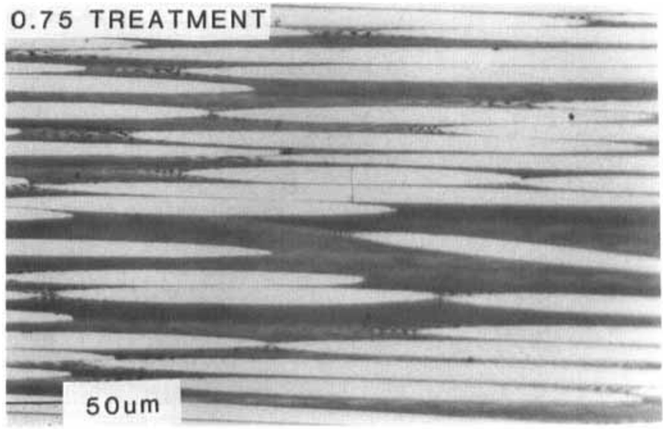

FIGURE 10 Hybrid sample showing two adjacent fibre breaks.

from the single fibre tests. At high surface treatment levels, composites were found to fail with the major crack running perpendicular to the fibre direction. This behaviour is well described by a model in which a crack grows from a critical accumulation of local fibre breaks. At lower surface treatments, longitudinal splitting was found to occur. As far as current models are concerned this could be considered as blunting of the growing i-plets, either before or after they become critical, so that the failure process is temporarily arrested. However, this is not accounted for in current models for failure. These failures of low surface treatment fibres extend across many transfer lengths so that application of a simple "chain of bundles" model is not appropriate.

Acoustic emission has been shown to have a direct relation to the expected number of single-fibre breaks in composite test-pieces; however, there is no apparent difference between the various surface treatments. This is to be studied in further detail.

The hybrid technique has been shown to be a useful way of studying failure mechanisms. The encapsulation of the impregnated tow into glass fibre/epoxy has little effect on their failure strains and Weibull parameter, but the failure zone is protected from consequential damage so that microstructural characterisation is possible.

More work is necessary in order to quantify the effect of surface treatment on the mechanisms of failure and its effect on the strength of the composites.

Acknowledgements

The work discussed in this paper is part of a more extensive investigation funded under the DTI-Link Structural Composites programme. The authors would like to thank their colleagues at British Aerospace, Sowerby Research Centre; Ciba-Geigy Bonded Structures, AKZO Fibres, The National Physical Laboratory and University of Bristol. We must also acknowledge Dr. Dick Biddulph for the development of the single fibre tensile testing apparatus, Anita Buxton for carrying out these single fibre tests and Reg Whattingham for his invaluable assistance at all stages of the project. Thanks also to Dr. Amir Rezaifard and Dr. Paul Smith for useful discussions within the department.

References

1. S. B. Batdorf, *J. Rein. Compos.* **1,** 153 (1982).
2. M. G. Bader, K. L. Pickering, A. Buxton, A. Rezaifard and P. A. Smith, *J. Fibre Sci. and Technol.* (1993), in press.
3. Private Communication—P. Marshall, BAe., Farnborough.
4. Private Communication—S. Plano and M. Kinsella, Akzo Faser AG.
5. G. F. Tudgey, RAE Farnborough Technical Report 91047 (1991).
6. P. W. Manders and M. G. Bader, *J. Mater. Sci.* **16,** 2233 (1981).
7. M. J. Pitkethly and M. G. Bader, *J. Appl. Phys.* **20,** 315 (1987).