Fibre fragmentation in multi-fibre micro and hybrid composites

Fibre fragmentation in multi-fibre micro and hybrid composites: The influence of fibre surface treatment.

Traineeship thesis by Wilco Verbeeten

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Fibre fragmentation in multi-fibre micro and hybrid composites: The influence of fibre surface treatment.

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Objective

The objective of the present report was to investigate the influence of fibre surface treatment on the failure process of multi-fibre microcomposites and carbon-glass hybrid composites.

Summary

Multi-fibre microcomposites were manufactured to investigate the failure mechanism in fibre reinforced materials. The influence of a fibre break on adjacent fibres was studied for both untreated and commercially treated carbon fibres. Compared to the treated fibres, untreated fibres show significantly more debonding, and the influence on adjacent fibres is almost none with these untreated fibres.

To investigate if the results found for the microcomposites are also valid for real composite structures, carbon-glass hybrid composites were manufactured and tested.

The results are similar. Higher surface treatment gives less debonding, smaller distances between fibre tips at fractures and a smaller positively affected length due to stress concentration of a broken adjacent fibre.

Contents

1. Introduction

The use of fibre reinforced composite materials is of a wide range. The last years the use has increased more and more. Its big advantage is that light weight is combined with high strength and high stiffness. Still, a lot of questionmarks can be put at the calculations of the mechanical behaviour of these fibre reinforced materials. Yet, there is not much known of failure processes and modes.

To understand the failure processes and modes of fibre reinforced composites, one has to look at the interface between the fibres and the matrix. To get a better insight one can first simplify things by using model composites. Quite a lot of research has been done earlier on investigating the interface strength of single fibres only. Recently, there has been a research project on single filament composites at the Eindhoven University of Technology [1]. Here, the model composites existed of one carbon fibre embedded in an epoxy matrix. Different surface-treated carbon fibres were used, such that different interface strengths were created.

The next step is to find out about the influence of surface treatment on the increased stress concentrations in the adjacent fibres. This can be done by using multi-fibre microcomposites. In a multi-fibre fragmentation test, an uni-axial load is imposed upon the sample, containing five parallel aligned fibres. The stress is transferred through the fibre-matrix interface to the fibre. If the stress gets to a certain level, fibre fracture occurs at the weakest point of the weakest fibre. Around the fracture, stress concentration occurs which leads to a higher stress level in the adjacent fibre.

In this report, we investigated the above described phenomena by testing multi-fibre microcomposites. Samples were made containing five carbon fibres embedded in an epoxy matrix and at three different distances between the fibres. The carbon fibres were not surface treated such that adhesion is on a low level. These samples were compared with the samples with commercially treated carbon fibres, *i.e.* 100%, to see the influence of the interface strength.

Regarding the adhesion between matrix and fibre and the stress build-up, it is to be expected that in multi-fibre microcomposites with 100% treated carbon fibres, bands of fractures can be detected. On the other hand, in the model composites in which 0% treated fibres are embedded, fibre failure will take place much more randomly (see Figure 1).

To investigate how the results obtained for multi-fibre microcomposites are to be translated to uni-directional composites, carbon-glass hybrid composites were manufactured and tested. In these samples carbon bundles impregnated with epoxy are surrounded by glass bundles also impregnated with epoxy. Here, the expectation is also that failure will occur more randomly in samples with 0% treated carbon. A larger difference between untreated and surface-treated carbon fibres can probably be seen at the distance between the two ends of the broken fibres. The higher the treatment, the smaller the distance between the fibre tips, because the adhesion with the epoxy is less and therefore relaxation of the fibres can take place.

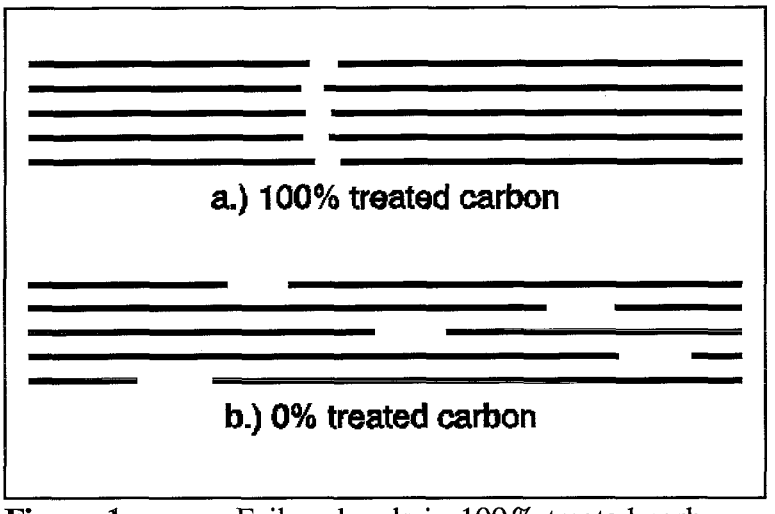

Figure 1: a. Failure bands in 100% treated carbon
b. Random fractures in 0% treated carbon

2. Experimental

2.1. Materials

The carbon fibres which were used for making the multi-fibre microcomposites and the carbon-glass hybrid composites, are unsized intermediate modulus carbon fibres (Courtaulds Grafil Apollo IM-44) containing 12000 fibres per yarn. The carbon fibres were oxidatively surface treated at different levels by the manufacturer, consisting of a very mild degree of etching in combination with the creation of active sites on the fibre surface for improving the adhesion [2,3]. The common commercial fibre treatment is known as 100%, the other treatment levels are named with respect to this treatment. For the multi fibre filaments the 0% and for the carbon-glass hybrids the 0%, 10%, 50%, 100% and 200% treated carbon fibres were used.

A combination of a common DGEBA type epoxy (Ciba Geigy, LY556) and 47 parts hardener per hundred parts resin triaminopoly (oxypropylene) curing agent (Texaco, Jeffamine T-403) was used. This epoxy system has the following required conditions: (i) it is transparent for following the fragmentation test visually; (ii) its viscosity is low during impregnation of the carbon fibres preventing premature fibre breakage; (iii) an almost linear stress-strain behaviour up to saturation of the fibre fragmentation process for modelling considerations; (iv) a low curing temperature for minimizing the effect of thermal shrinkage [4].

For preparing the carbon-glass hybrid composites E-glass fibres was needed. These E-glass fibres must have a higher strain-to-break than the carbon fibres for preventing the hybrid composite samples from premature breaking. The E-glass chosen was Metha-Epox (Silenka, 084-M28) containing 1200 fibres per yarn.

2.2. Sample manufacturing

2.2.1. Multi-fibre microcomposites

A tensile bar containing five carbon fibres was manufactured using a fibre positioner, developed by Wagner and Steenbakkers [5], and a dogbone shaped silicone rubber mould (see Figure 2 and 3).

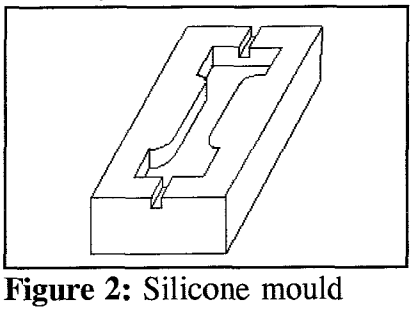

Figure 2: Silicone mould

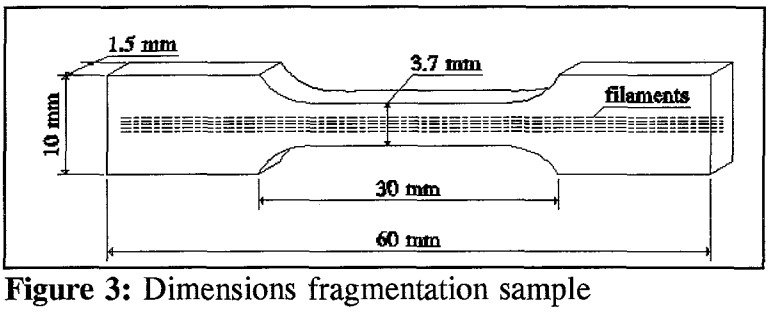

Figure 3: Dimensions fragmentation sample

The fibre positioner is shown in Figure 4. The fibres are positioned between cylindrical rods, having a centre-to-centre distance of 550 μm. The fibre positioner has a micrometer, for installing an angle Θ, with which the centre-to-centre distance between two adjacent fibres can be reduced to $550 \cdot \cos(\Theta)$ μm (see Figure 5).

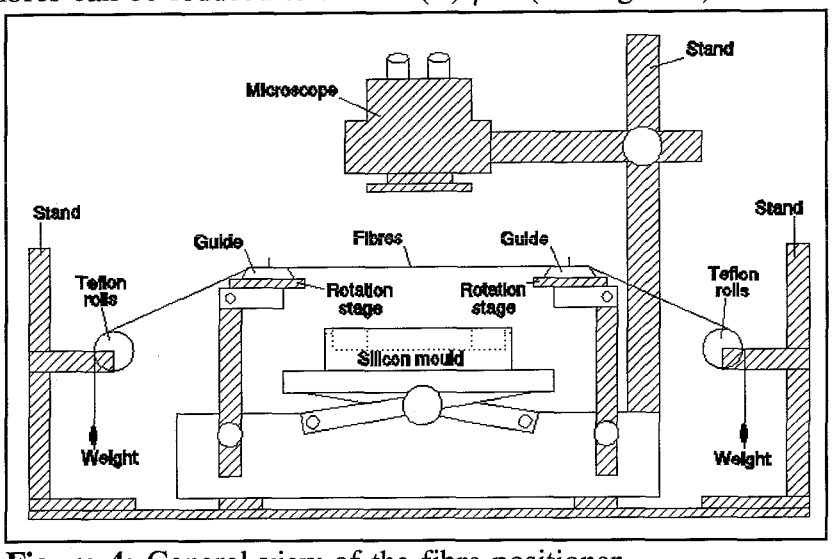

Figure 4: General view of the fibre positioner

Five fibres were picked randomly from a carbon bundle and placed on the positioner. Three inter-fibre spacings were made, namely a fibre-fibre distance of 3 fibre diameters (equal to 14.7 μm), 9 fibre diameters (equal to 44.1 μm) and a fibre-fibre distance of 20 fibre diameters (equal to 98.0 μm). On both ends of all five fibres, photo stickers were attached having a total weight per fibre of 0.140g. For all fibres, the weight was equal to exclude the influence of different fibre pre-tension on the fragmentation test results [6]. Next, the silicone mould was positioned and the fibres were fixed on the mould with fast curing glue (UHU). After curing the glue, the degassed (60°C, 20 min, 400Torr) epoxy resin was injected carefully with a syringe. Then, a glass plate provided with release coating (Hysol, Frekote) and a weight of 3kg were put on the mould. The samples were cured at Room Temperature for 24h, followed by 16h at 75°C.

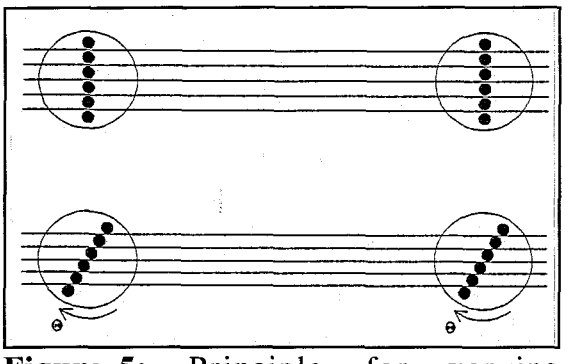

Figure 5: Principle for varying fibre-to-fibre distance

2.2.2. Carbon-glass hybrid composites

To make sure the hybrid composites do not break after failure of the carbon bundle, the volume fraction of the E-glass has to be more than a critical value. This critical volume fraction can be determined with Formulas 1 and 2, based on the 'constant strain' model in hybrid composites [7]. The formulas were derived from the strength diagram which is shown in Figure 6. The strength of the hybrid is given by the two straight lines AC and CD, were point A denotes the tensile strength of the carbon composite and point D that of the glass composite.

$$\sigma_{hybrid} = \sigma_{carbon,max} \cdot V_{carbon} + \epsilon_{carbon,max} \cdot E_{glass} \cdot V_{glass} \qquad \forall\ V_{glass} < V_{crit} \tag{1}$$

$$\sigma_{hybrid} = \sigma_{glass,max} \cdot V_{glass} \qquad \forall\ V_{glass} > V_{crit} \tag{2}$$

$E_{carbon,impregn.}$	175 [GPa]
$\epsilon_{carbon,impregn.}$	1.8 [%]
$\sigma_{carbon,impregn.}$	2.9 [GPa]
$E_{glass,impregn.}$	35 [GPa]
$\epsilon_{glass,impregn.}$	2.6 [%]
$\sigma_{glass,impregn.}$	1.0 [GPa]

Table 1: Material properties by Courtaulds [8] and Silenka [9]

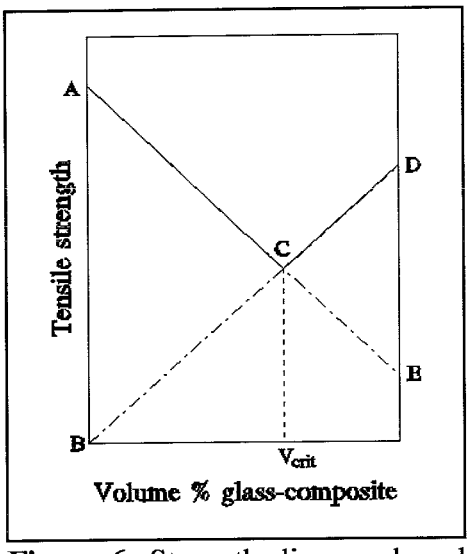

Figure 6: Strength diagram based on constant strain model for hybrid composites

With the contents of Table 1, one can determine that $V_{crit} = 0.89$. With a glass bundle density of approximately 13 bundles/cm and a sample width of ± 10 mm the glass volume fraction is over 90 percent.

The carbon-glass hybrids were manufactured using a filament winding machine. A window shaped framework was put into the winding machine. First, up to seven epoxy impregnated carbon bundles were distributed on the frame with equal spacing. Then, impregnated E-glass was wound onto the framework (see Figure 7). The framework was put in a metal mould between two pieces of Mylar foil and two brass plates all provided with release coating (Hysol, Frekote). Pressure was applied for an hour at 20 kN and 75°C. The composite was postcured for 15h at 75°C. Finally, the samples were cut out of the sample plate. Four small metal plates were attached at the ends of the samples for better pressure distribution during clamping in the tensile machine. The glue used for attaching is an epoxy glue based on Araldit AW 106 and HV 953 U. Dimensions of the samples are shown in Figure 8.

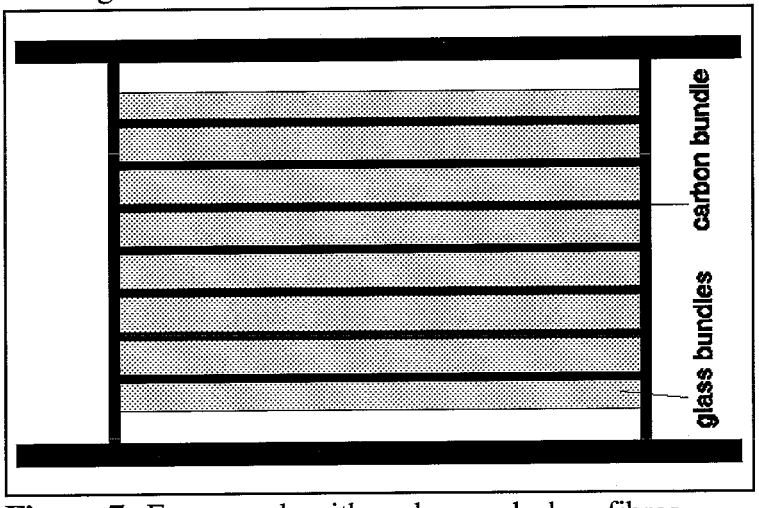

Figure 7: Framework with carbon and glass fibres

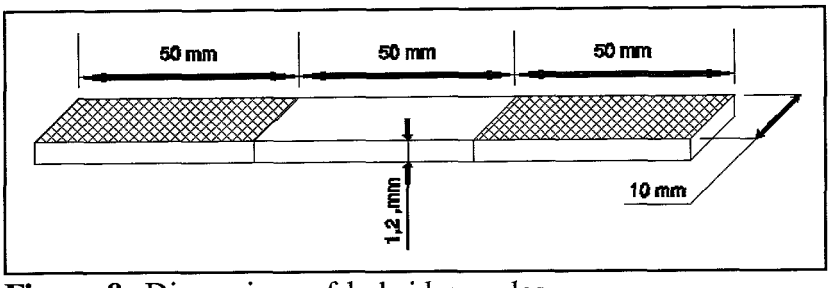

Figure 8: Dimensions of hybrid samples

2.3. Testing

2.3.1. Multi fibre model composites

Fragmentation tests on the multi fibre model composites were performed in two different ways. First, the acoustic emission technique (Physical Acoustics Corporation, Locan AT) was used to determine saturation of the fragmentation test. The fibre failure produces a pressure wave in the specimen which can be detected with a piezoelectric sensor.
To improve the sound transfer, grease was provided between the sensor and the sample. The samples were tested on a Frank 81565 tensile machine with a load cell of 10 kN at a loading speed of 0.01 % ϵ/sec (=0.2 mm/min) for each sample. The fragmentation tests were stopped when no more hits were detected. The tested samples were investigated using a polarized-light Zeiss microscope. They were checked for the real amount of fibre breaks and for measuring the fragment lengths of the fibres.
Secondly, the failure mode was determined using a small hand driven tensile machine. The samples were strained while placed under a polarized-light Zeiss microscope and recorded on video and photographs.

2.3.2. Carbon-glass hybrid composites

The hybrid samples were tested on a Zwick 1484 200kN load cell tensile machine at a loading speed of 0.01 % ϵ/sec (=0.3 mm/min). It was possible to visually determine the end of these fragmentation test.
After testing the hybrid samples, a longitudinal section was taken of the carbon bundle and examined under an Olympus microscope.

3. Results and discussion

3.1. Multi-fibre microcomposites

FRAGMENTATION PROCESS

Under a Zeiss microscope equipped with polarizers, the stress build-up process can be visualized. Figure 9 shows the fragmentation process of the 0% treated carbon fibres at a distance of 9 diameters. After rupture, the stress build-up around the fibre tip is very small. Also, the fibres are debonded over a long distance. This can best be seen in the first picture of Figure 9. The brightest spots are the places where the matrix and fibre are still attached. Between these spots are the debonded zones. When the load is increased, the debonded zones of the already broken fibres becomes even bigger, in a stick-slip kind of way. This stick-slip behaviour is also detected by the Acoustic Emission technique. During the fragmentation process, a lot of hits were detected but a lot less were counted when the sample was checked under the Zeiss microscope. In the Amplitude versus Time plot shown in Figure 10, two clouds can be seen. The upper cloud indicates the breaking of fibres, the one at 65dB is caused by the friction of the fibre and the matrix in the debonded parts.

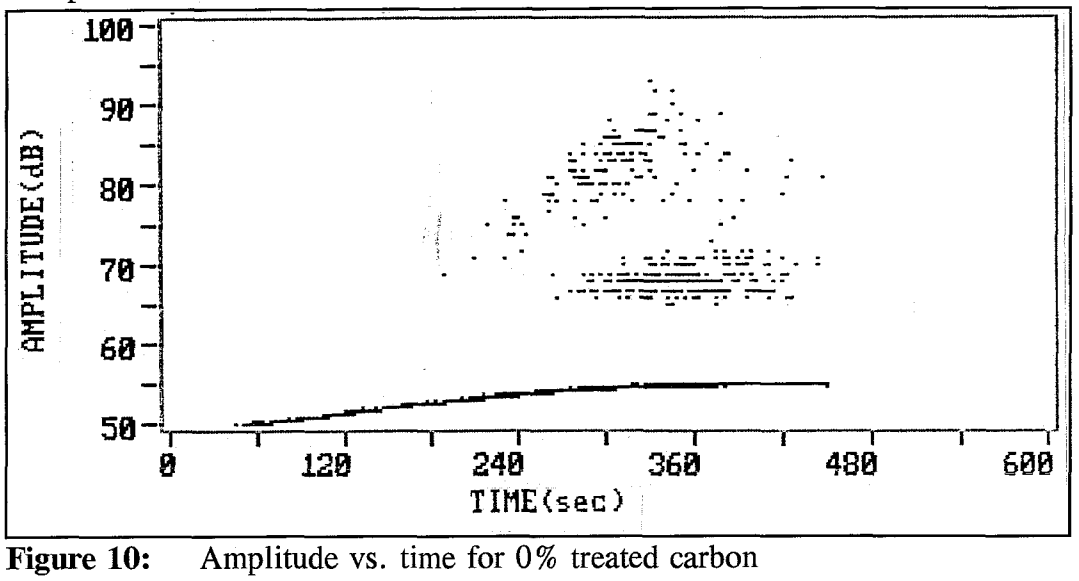

Figure 10: Amplitude vs. time for 0% treated carbon
Curve: Load vs. time

The sequential pictures in Figure 9 indicate that there is almost no influence of a broken fibre on an adjacent fibre. The fibre fractures randomly appear at different positions. No fractures occur at the same place in an adjacent fibre at the same time.
The last picture of Figure 9 shows the saturated sample. This is called saturated because the fibre fragment lengths are too short to build up enough stress for another fracture.

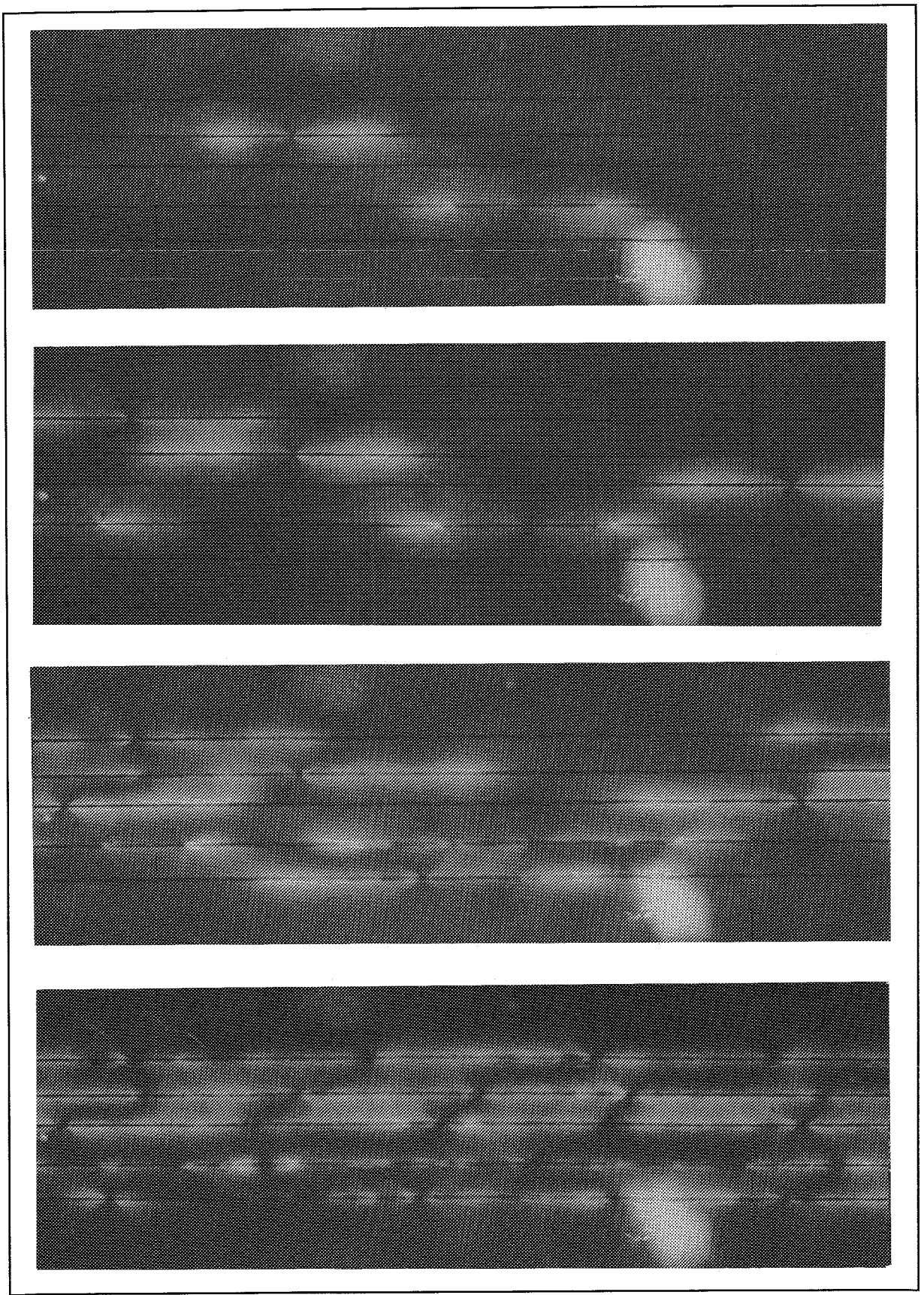

Figure 9: Stress pattern in sample with five 0% treated carbon fibres at 9 diameters distance

INFLUENCE OF SURFACE TREATMENT

Figure 11 shows two pictures of saturated samples containing five carbon fibres at a distance of 3 diameters. In the upper one 0%, and in the lower one 100% treated carbon is used. Here, it can be easily seen that the 0% treated carbon sample is fractured randomly while the 100% treated carbon has broken in fracture bands. The light sheaths in 0% treated carbon are longer and less bright. This is a result of the longer debonded zones and the lower stress build-up which is explained by a deterioration of adhesion between the fibre and matrix.

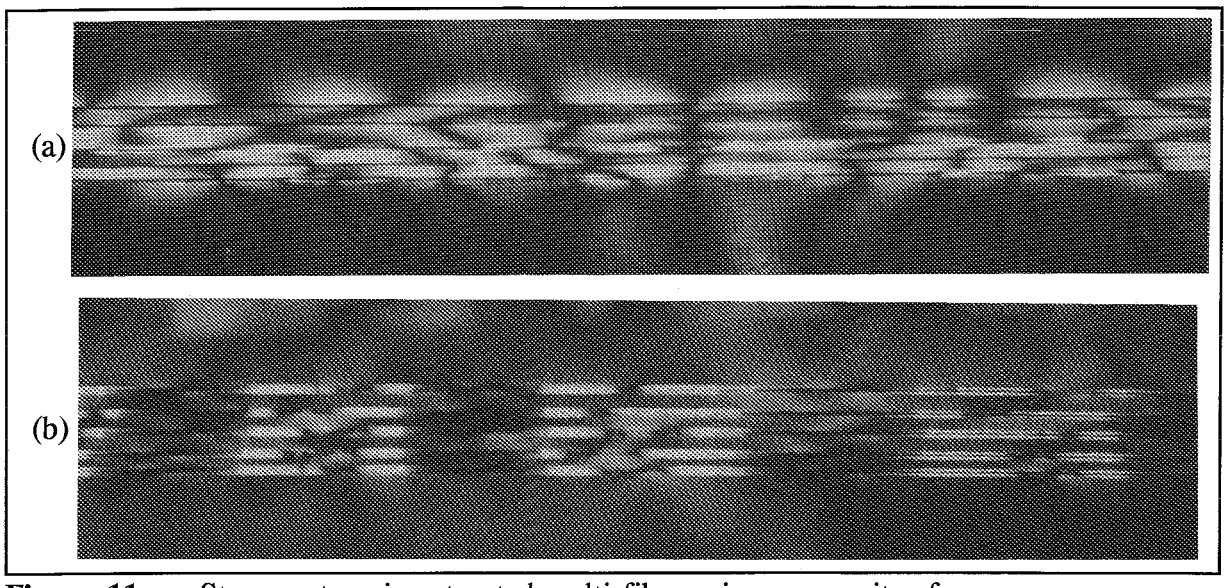

Figure 11: Stress patern in saturated multi-fibre microcomposites for:
(a) 0% treated carbon (b) 100% treated carbon

When the adhesion of the fibre to the matrix is low, *i.e.* 0% treated fibres, debonding occurs around the fibre break. Consequently, the stress level in the adjacent fibre is not increased very much and fibre fracture is not likely to take place.
When adhesion is on a higher level, *i.e.* 100% treated fibres, stress concentration will occur around the fibre fracture tip. This leads to a higher stress level in the adjacent fibre and failure is more likely to take place (see Figure 12).

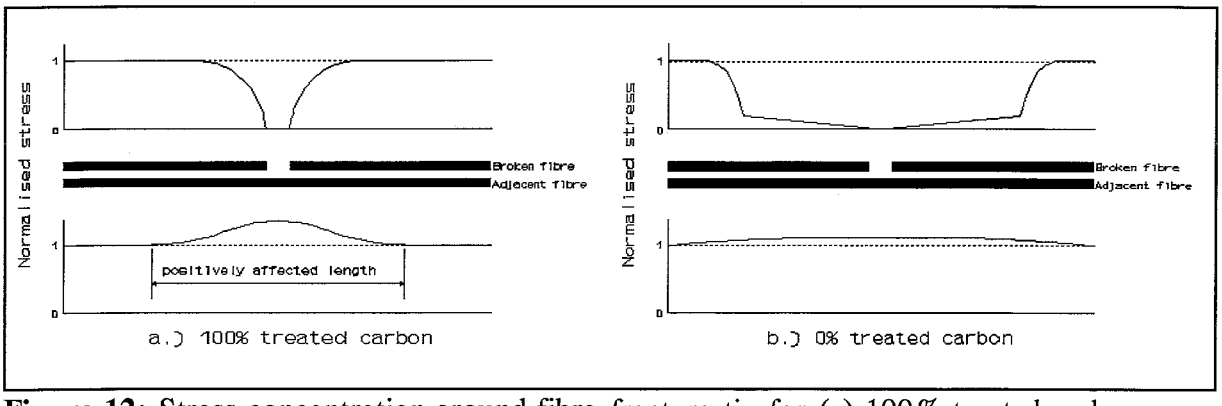

Figure 12: Stress concentration around fibre fracture tip for (a) 100% treated and (b) 0% treated carbon

Under a Zeiss microscope, a few saturated fragmentation samples with 0% and 100% treated carbon fibres were investigated and the positions of fractures were registered. Afterwards, a computer programme counted the number of breaks at the same position in adjacent fibres. The results are shown in Figure 13. It is shown that the visual obervations are confirmed: 0% treated carbon samples have a broader, random distribution and in the case of 100% treated carbon samples almost all fractures occur in bands.

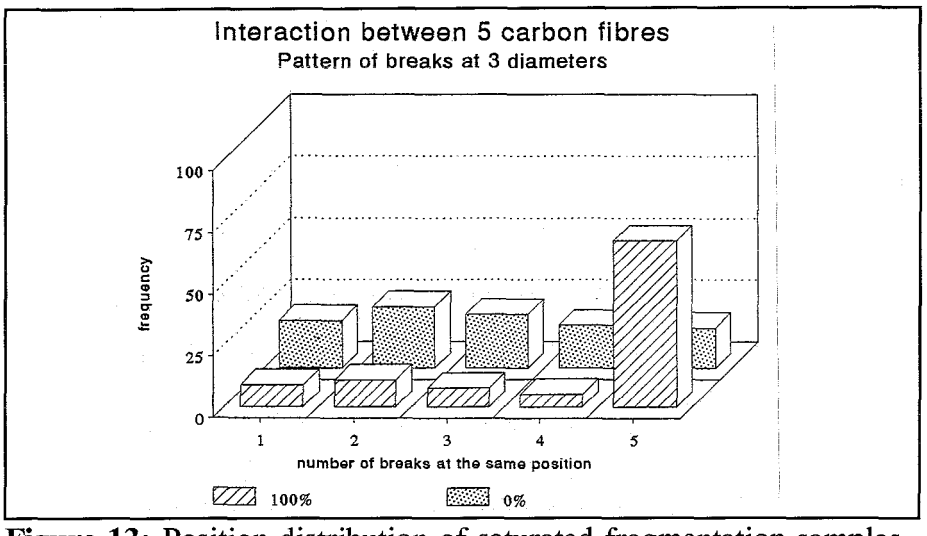

Figure 13: Position distribution of saturated fragmentation samples

INFLUENCE OF FIBRE SPACING

When we compare the samples with 0% treated carbon fibres at the three different distances, almost no difference can be detected (see Figure 14). Even at a distance of 3 diameters there are almost no bands. The influence of stress on an adjacent fibre is only insignifanctly present at the distance of 3 diameters. Figure 15 shows a slightly higher frequency of five breaks at the same position at a distance of 3 diameters. Between the graphs of 9 and 20 diameters the difference is almost absent. These two graphs have the same distribution and almost the same frequency at all number of breaks.

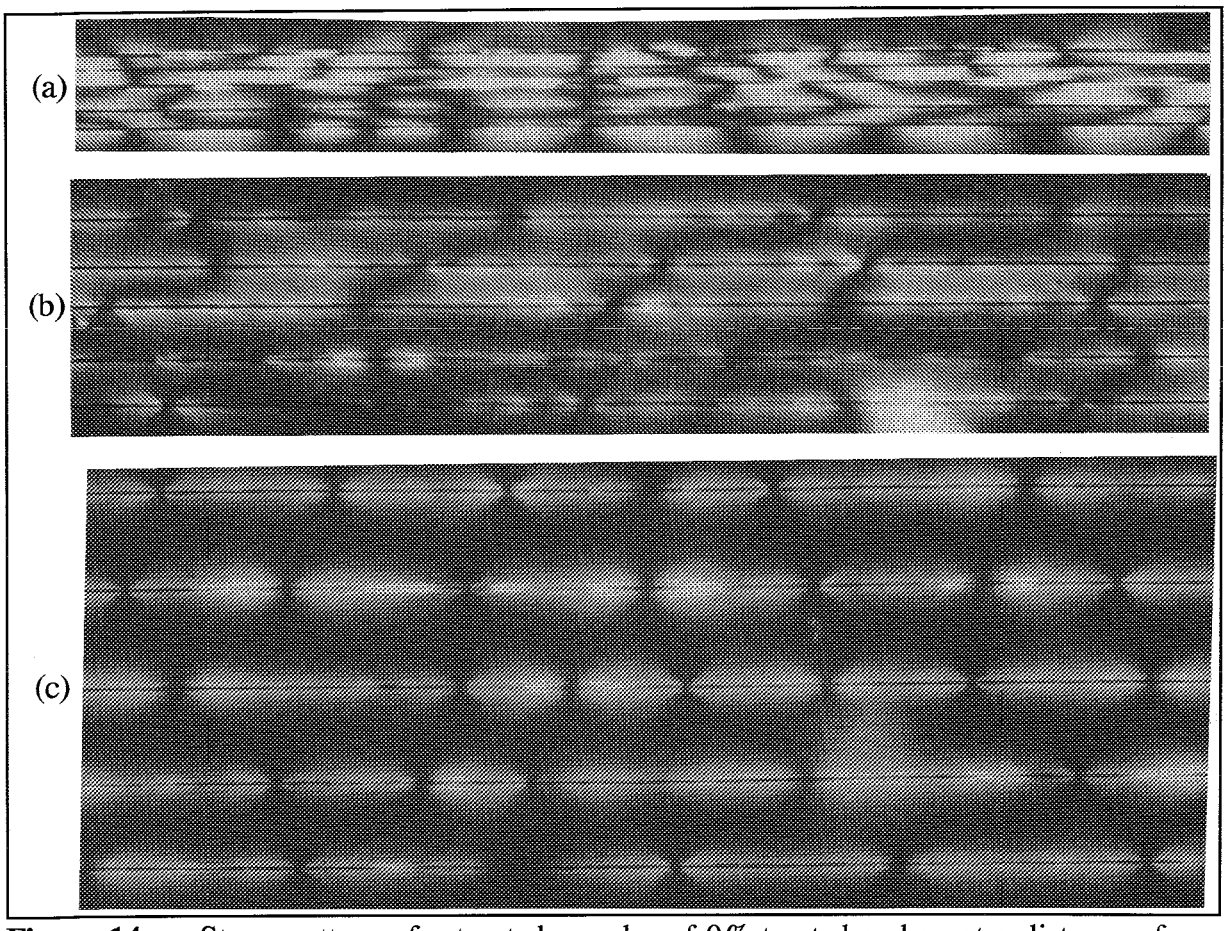

Figure 14: Stress pattern of saturated samples of 0% treated carbon at a distance of: (a) 3 diameters (b) 9 diameters (c) 20 diameters

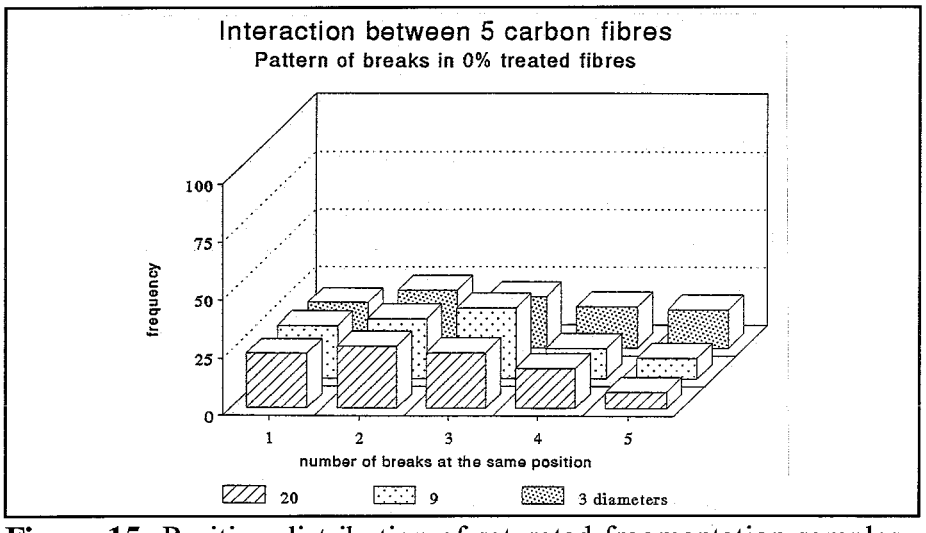

Figure 15: Position distribution of saturated fragmentation samples

3.2. Carbon-glass hybrid composites

All hybrid samples were strained to the point that delamination occured at two or more places. This delamination was accompanied by a sudden, audible event.
General features of the failure mode of the samples are shown in the photographs of Figure 16. As the level of fibre surface treatment is increased, the failure zone, in the longitudinal sense, is smaller. Except for 50% treated carbon, which has coincidentally the smallest zone. Also, the distance between the two fibre tips gets smaller at higher treatment levels. At each level of surface-treatment, close to the bundle failure, a zone of debonding is observed to form between the carbon bundle and the glass/epoxy 'matrix'. These above-mentioned effects were also reported by Bader [10]. A higher level of surface-treatment reduces the positively affected length, due to the stress concentration of an adjacent broken fibre, and increases the stress concentration. It also results in a better adhesion between fibres and 'matrix' and therefore prevents broken fibres from relaxation. At low treatment levels adjacent fibre breaks are further apart and shear failure spreads the failure zone along a greater length of the bundle.
These effects were also observed at the multi-fibre microcomposites.
Another effect also reported by Bader, can be seen in Figure 16(c). Here, the fracture path has forked. It would appear that the failure originates towards the centre of the bundle and propagates outwards. The debonding between bundle and 'matrix' appears to run outwards from the two fork segments, leaving a still-bonded region in between.

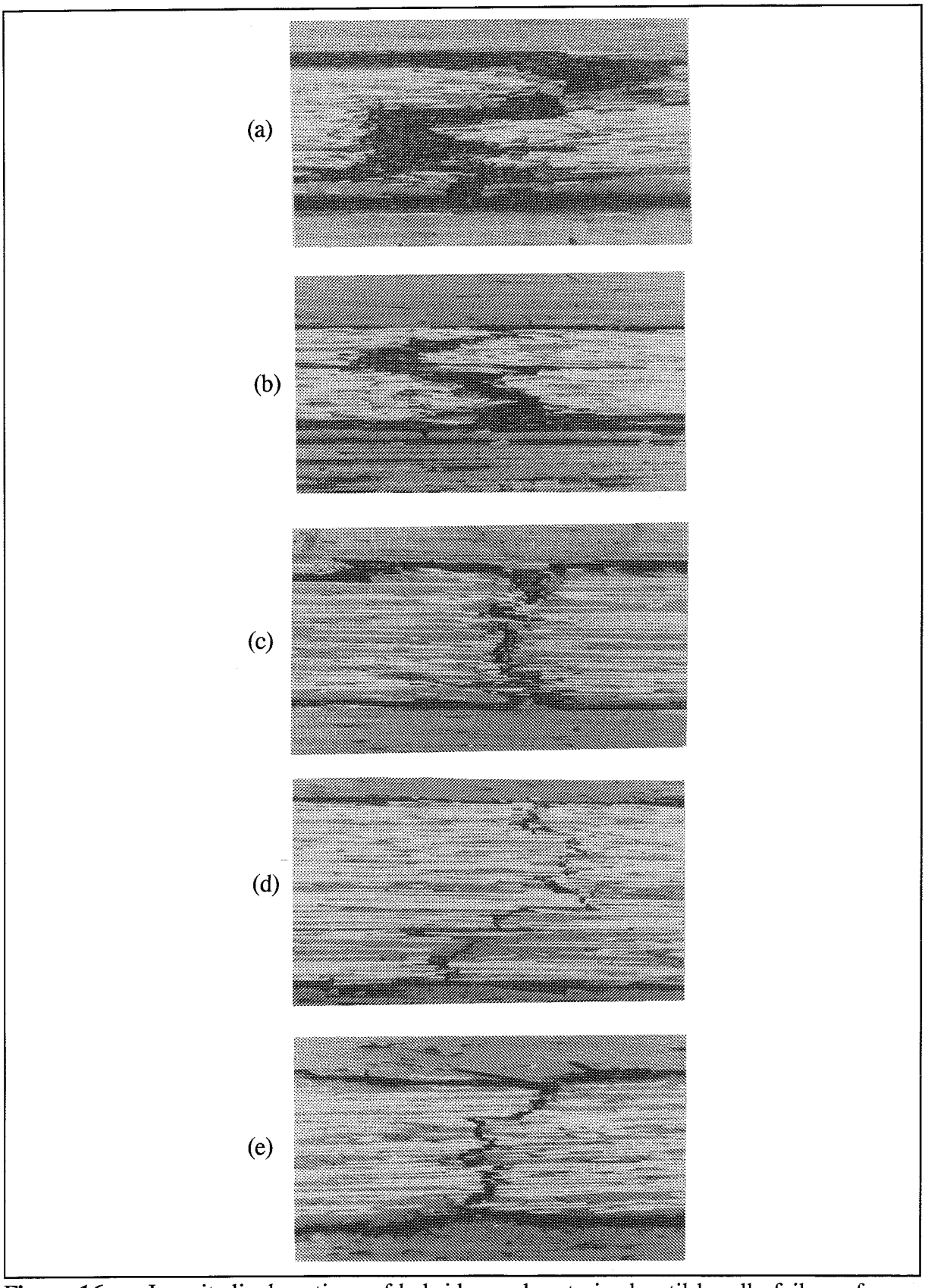

Figure 16: Longitudinal sections of hybrid samples strained until bundle failures for: (a) 0%, (b) 10%, (c) 50%, (d) 100% and (e) 200% treated carbon. Magnification: 10,000×

4. Conclusions and recommendations

The multi-fibre microcomposites have given a better insight into the failure mechanisms of composite materials. With videorecordings and microphotographs, the failure phenomena, as debonding and stress build-up, could satisfactorily be visualized. The influence of different treatment levels was clearly visible. The influence of untreated carbon fibres on each other was almost negligible. Only at a distance of 3 diameters a little influence could be detected. The debonding zones of untreated carbon fibres were much bigger than at higher treated fibres. Also, the fragments were longer. Stress build-up was spread over a bigger zone and was less high than at 100% treated carbon.

For the carbon-glass hybrid composites a similar result could be detected. At a higher treatment level of the carbon bundles, the positively affected length, due to the stress concentration of a broken fibre, was shorter. Therefore, the breaks in adjacent fibres were closer together and resulted in a failure path more perpendicular to the axis of the bundle. Furthermore, the higher the level of treatment, the smaller the fragments. This was also observed at the multi-fibre microcomposites.

In every sample and at every level of treatment, near the main bundle failure debonding occured between the bundle and the glass/epoxy 'matrix'.

While the hybrid composites were only viewed after failure, a better understanding of the failure mechanism can be obtained by following the complete failure process with a microscope if possible and an extensometer.

5. References

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