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Study of the interlaminar shear strength of unidirectional high-modulus polyethylene fibre composites

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The interlaminar shear strength of a unidirectional high-modulus polyethylene fibre–epoxy resin composite, measured using a short-beam bend test, increases when the fibre has been plasma treated but decreases with increasing fibre content or increasing filament diameter. Attempts have been made to explain these observations. From observation of the sample failure and the bending load/deflection curves, it was found that there are two possible failure modes dependent on fibre type, fibre volume fraction and fibre plasma treatment. These are (1) brittle failure of the resin and (2) failure at the fibre/resin surface. The present results appear to be consistent with the interlaminar shear strength data obtained from high-modulus polyethylene fibre samples made by the hot compaction process developed at Leeds.

1. Introduction

The effect of fibre volume fraction on the unidirectional bending failure stress of high-modulus polyethylene (HMPE) fibre composites was studied.

The high specific strength and modulus of HMPE fibres can be utilized to produce a fibre composite with a high specific strength and modulus. The flexural and impact properties of the HMPE fibre composite will be dependent on the adhesion between the fibre and the resin, which will determine both the flexural failure strain and the impact energy absorption [1]. The composite behaviour during deformation will, therefore, depend on the relative shear behaviour of the fibre and the resin matrix together with the adhesion between the fibre and the matrix. Thus, although the flexural modulus of a composite containing high-strength fibre will increase with increasing fibre content, the properties such as flexural strength, which depend on the shear properties of each material, will not necessarily similarly increase.

2. Experimental procedure

2.1. Materials

2.1.1. Fibre

The HMPE fibres were produced on a commercial scale using either a melt-spinning process or a gel-spinning process followed by hot drawing. In addition, HMPE multifilaments, and also monofilaments having a considerably larger diameter, were produced by melt spinning and drawing in the laboratory from a low molecular weight polymer. Details of the fibre properties and their origins are given in Table I.

2.1.2. Resin

Epoxy resin (Ciba Geigy LY 1927) was used as the matrix in the unidirectional composites. This is a low-viscosity resin intended for high-strength composites with a bending modulus of 2.5 GPa for the unfilled cured resin.

2.2. Preparation of unidirectional fibre/epoxy resin composites

Unidirectional composites measuring 200 mm × 10 mm × 2 mm were prepared using all the fibres listed in Table I. A leaky mould technique was employed as described in a previous paper [2].

2.3. Preparation of compacted unidirectional HMPE fibre samples

Samples of compacted unidirectional HMPE fibre with dimensions 200 mm × 10 mm × 2 mm were made from all of the fibres listed in Table I using a hot compaction process developed at Leeds [3], which is the subject of a patent application [4]. In this process, a unidirectional bundle of untreated HMPE fibres is held in position under a comparatively low pressure (approximately 500 p.s.i.; 1 p.s.i. = 6.89×10^3 N m^{-2}) and heated to an accurately defined temperature close to the crystal melting temperature. After application of a high pressure (approximately 9000 p.s.i.) for a short time, the sample is cooled. During the compaction process, a surface layer of each filament (approximately 0.08 volume fraction) is melted and flows into the voids between each filament. During cooling, this melted material crystallizes in an unoriented form and makes only a small subsequent contribution to the longitudinal strength. However, the bulk of the filament is unaffected by this compaction process and the compacted sample has a density and a bending modulus corresponding closely to the properties of the original HMPE fibre (Table II). It can therefore be

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TABLE I Fibre properties

Fibre type	Fibre origin	Filament diameter (mm)	Fibre strength (GPa)	Elongation (%)	Initial modulus (GPa)	Molecular weight, M_w ($\times 10^3$)
Experimental multifilament	Leeds	0.015	1.0	3.3	54	61
Tenfor	Snia Fibre, Italy	0.013	1.3	4.7	57	130
Experimental monofilament	Leeds	0.055	0.9	3.3	41	61
Tekmilon	Mitsui, Japan	0.038	1.8	2.8	80	700
Dyneema	DSM, Holland	0.012	2.6	3.5	87	1300
Spectra 1000	Allied-Signal, USA	0.027	2.7	2.4	120	1500

TABLE II Compacted unidirectional fibre composites

Fibre	Compacting temperature	Compacted fibre bending modulus	Fibre tensile modulus	Compacted fibre ILSS	Extrapolated ILSS from Fig. 3	Extrapolated ILSS from Fig. 2
	(°C)	(GPa)	(GPa)	(MPa)	(MPa)	(MPa)
Multifilament	133.9	27	54	13.5	20	5
Tenfor	137.4	28	57	17.2	17	5
Tekmilon	146.2	31	80	11.7	25	13
Dyneema	149.0	39	87	13.6	15	7
Spectra	149.0	54	120	14.9	13	5

considered as a unidirectional HMPE fibre/polyethylene composite with a fibre volume fraction of approximately 0.92. Attempts to compact the much larger diameter monofilaments were unsuccessful, possibly because of insufficient available pressure.

2.4. Plasma treatment

The fibres were plasma treated in a batch process using an oxygen gas plasma oven as described previously [5]. For each treatment, a total of approximately 6 g fibre were treated for 2 min at a power of 120 W before incorporation in the unidirectional composite. These conditions were selected to give a relatively large increase in adhesion with a minimum of strength loss.

2.5. Interlaminar shear strength testing

The adhesion between the fibre and the resin matrix was measured using a short-beam bend test with a sample thickness of 2 mm and a bending span-to-sample thickness ratio of 5:1 which is sufficiently low to produce failure in the shear mode. A 20 mm × 10 mm × 2 mm unidirectional composite sample was flexed between 6 mm diameter support rods with a centre distance of 10 mm and a central 6 mm diameter rod moving at 2 mm min^{-1}. The peak load, F (kg), was used to calculate the shear stress along the mid-plane or the interlaminar shear strength (ILSS).

For the sample of width w (mm) and thickness t (mm)

$$\text{ILSS} = \frac{7.357F}{wt}\,\text{MPa} \qquad (1)$$

Each result is the mean of four short-beam flexural tests made on samples cut from one unidirectional composite, with a standard error in the mean of 0.5 MPa.

3. Results

Fig. 1 shows that the shear failure stress (ILSS) of unidirectional melt-spun HMPE (Tenfor) untreated fibre composites calculated from the peak bending load observed during a short-beam bend test decreases as the fibre volume fraction increases. At low volume fractions, the samples exhibit brittle failure. A similar dependence of ILSS on fibre volume fraction is also seen in Fig. 1 for composites containing plasma-treated Tenfor fibre, the higher ILSS values at each fibre volume fraction reflecting the increase in adhesion produced by plasma treatment.

A similar result is obtained in Figs 2 and 3 for the gel-spun Spectra and Dyneema fibres and the melt spun Tekmilon and Multifilament fibres. It is also seen in Figs 1–3 that the shear failure data for all of the fibres appear to extrapolate to 59 MPa at zero volume fraction. At high volume fraction, the plasma-treated data extrapolate to shear failure values in the region of 15–25 MPa. In contrast, most of the untreated fibre data extrapolate to values in the region of 5 MPa. Table II compares these extrapolated values of ILSS for plasma-treated fibre at a fibre volume fraction of 0.92 with the ILSS values obtained for compacted fibre.

A comparison between samples produced from melt-spun fibres of differing diameter in Fig. 4 shows that for both untreated and plasma-treated samples, the shear failure stress is lower for the larger diameter fibre. As with the other fibres, the shear failure data for the monofilament extrapolates to 59 MPa at zero volume fraction. Also, at low fibre volume fraction, the failure is again in the brittle mode and the failure stress

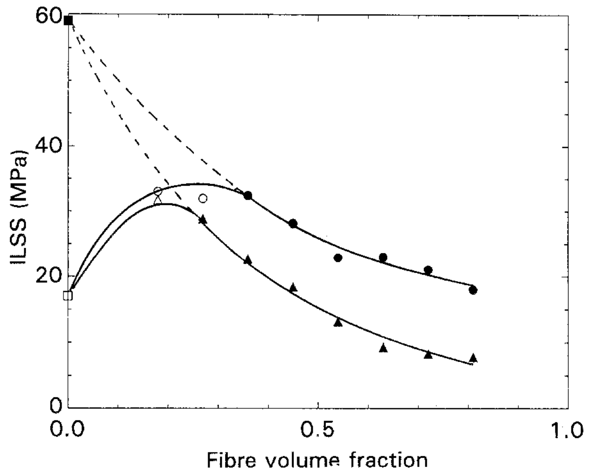

Figure 1 Effect of fibre volume fraction and plasma treatment on ILSS, Tenfor fibre: (△, ▲) untreated fibre, (○, ●) plasma-treated fibre, (▲, ●) shear failure, (△, ○) brittle failure.

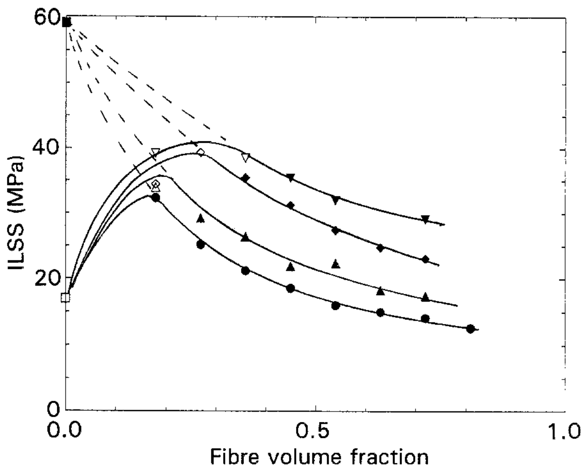

Figure 3 Effect of fibre volume on ILSS for plasma treated fibre: (●) Spectra, (▽, ▼) Tekmilon, (◇, ◆) multifilament, (△, ▲) Dyneema, (□, ■) resin only, (●, ▼, ◆, ▲) shear, (▽, ◇, △, □) brittle failure.

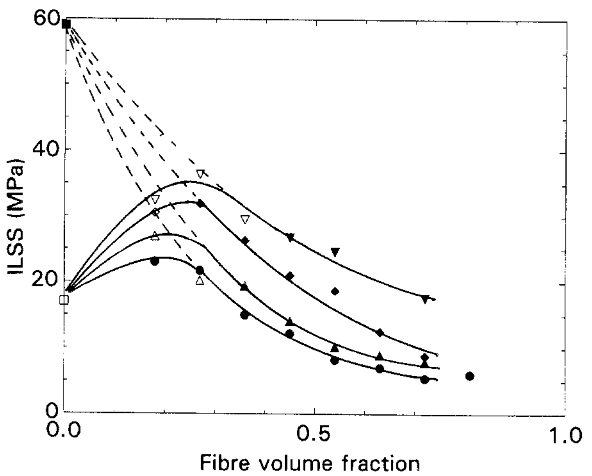

Figure 2 Effect of fibre volume fraction on ILSS for untreated fibre: (●) Spectra, (▽, ▼) Tekmilon, (◇, ◆) multifilament, (△, ▲) Dyneema, (□, ■) resin only, (●, ▼, ◆, ▲, ■) shear failure, (▽, ◇, △, □) brittle failure.

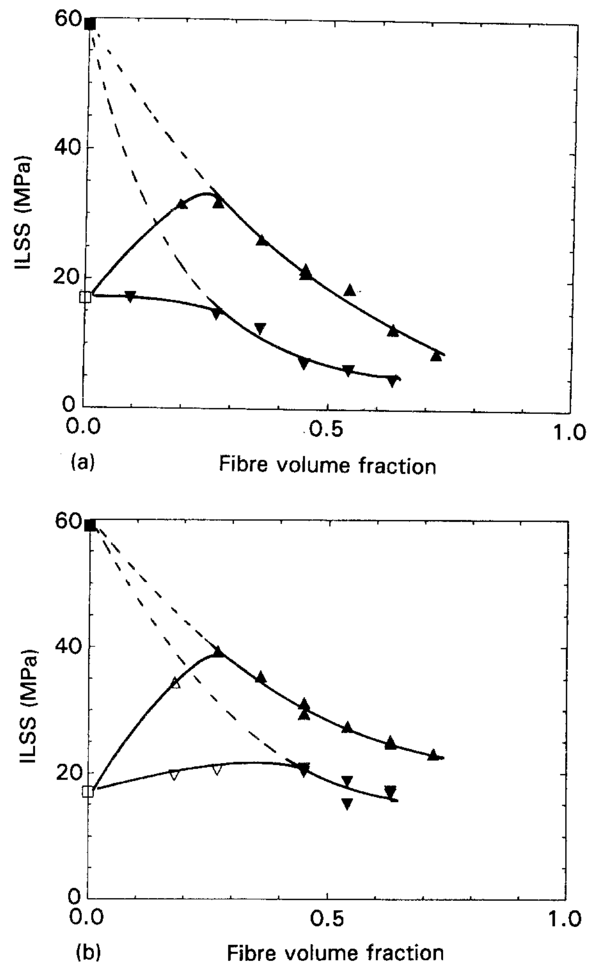

Figure 4 Effect of filament diameter on ILSS: (a) untreated fibre, (b) plasma treated fibre, (△, ▲) 0.015 mm multifilament, (▽, ▼) 0.055 mm monofilament, (▲, ▼) shear failure, (△, ▽) brittle failure.

values are reducing and approaching the resin failure value at zero fibre content.

The reinforcement of resin with fibre to form a composite will produce a progressive increase in tensile properties, the increase following the rule of mixtures. In particular, the bending modulus, which is proportional to the tensile modulus, will be proportional to the fibre volume fraction. Measurements made with the multifilament fibre unidirectional composites are shown in Fig. 5 and confirm that this relationship does apply for these composites. A particular feature of this result is that both the unfilled resin at zero volume fraction and the unidirectional compacted fibre results at 0.92 fibre volume fraction are consistent with the unidirectional fibre-reinforced resin composite results.

To obtain a measure of the shear failure stress of the epoxy resin used in this work, cylinders of the resin were moulded and compression tested on the Instron tester. Using cylinders having a length/diameter/aspect ratio of 11.9 mm/7.9 mm/1.5, 15.2 mm/7.6 mm/2.0 and 21.8 mm/10.9 mm/2.0 respectively, and a compression rate of 10 mm min^{-1}, the failure was ductile and a compression failure stress of 118 MPa was obtained for each sample. Assuming that the yield stress is independent of pressure, this gives a shear failure stress of 59 MPa for the resin.

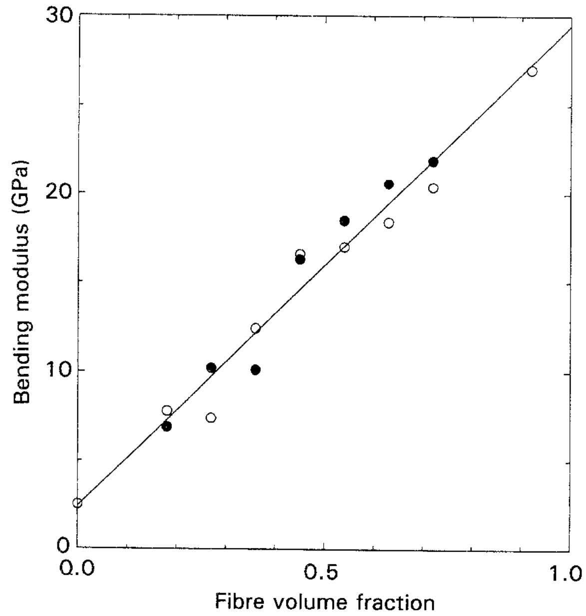

Figure 5 Bending modulus of multifilament fibre composites: (●) untreated fibre, (○) plasma treated fibre.

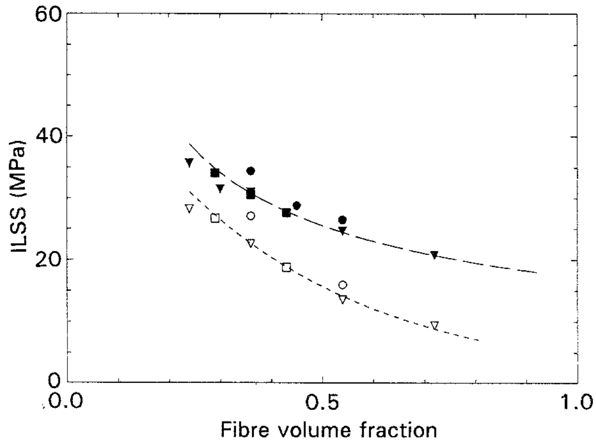

Figure 6 Effect of composite thickness on ILSS, Tenfor fibre: (○, ●) 2 mm, (□, ■) 2.5 mm, (▽, ▼) 3 mm, (●, ■, ▼) plasma treated, (○, □, ▽) untreated.

4. Discussion

A measure of the adhesion between the reinforcing fibre and the resin matrix in a unidirectional composite can be obtained from a short-beam bend test [2]. The peak load from this test is used to calculate the interlaminar shear strength (ILSS) of the composite. The results obtained have shown that the ILSS values obtained depend not only on the HMPE fibre type but also on fibre diameter, fibre plasma treatment and fibre volume fraction of the composite: the ILSS increases with decreasing diameter, decreasing fibre volume fraction and with plasma treatment.

Previous work by Ladizesky and Chow [6] has indicated that an increase in Tenfor fibre volume fraction in a unidirectional composite from 0.48 to 0.60 produces a small reduction in ILSS. They also observed a similar reduction in ILSS after the sample thickness was reduced from 2.5 mm to 2 mm. The present studies confirm the volume fraction effect, although no effect of sample thickness has been found: Fig. 6 shows that 2, 2.5 and 3 mm thick samples containing fibre volume fractions of 0.24–0.54 for both untreated and plasma-treated Tenfor HMPE fibre gave ILSS results, at a constant test span/thickness ratio of 5:1, in good agreement with the data obtained in Fig. 1 for 2 mm thick samples at the same test aspect ratio.

To attempt an understanding of the failure mechanisms, both the appearance of the samples before and after failure and the shape of the bending load/deflection curves have been studied. Also, both unreinforced resin samples and samples made by compaction were included in the study to represent the extremes of fibre concentration.

Observation of the failed samples containing Tenfor in Fig. 1, show (1) brittle failure at low fibre volume fraction, and (2) shear failure at higher fibre volume fraction. Similar findings were obtained for the other fibres in Figs 2–4 and it is concluded that these two failure mechanisms determine the failure mode depending on the amount of fibre present. This is confirmed by results in Fig. 7 which shows photographs of the samples that have been deflected just beyond their failure strain. Fig. 7a–c show that for low fibre content and also for resin only, the samples exhibit brittle failure, with no permanent deformation either side of the central failure point. At high fibre content and also with the compacted fibre sample, Fig. 7d–f show that only shear failure has occurred, resulting in permanent deformation of the composite between the beam supports. Observations of samples made using the other fibres in Table I yield identical conclusions.

It is of particular interest to note in Table II that, apart from the Tekmilon fibre, the other four compacted samples give ILSS values that are close to the extrapolated values for the plasma-treated composite data in Fig. 3. Also, they are considerably greater than the corresponding extrapolated values of ILSS for the untreated composite results in Fig. 2. Although the compacted fibres are made using untreated fibres, the filament surfaces are adhered by the melted unoriented polyethylene that fills the gaps between the filaments, producing good adhesion between the highly oriented filaments and the unoriented polyethylene that links the filaments. It therefore appears that for high volume fractions of plasma treated fibre in a unidirectional composite, the failure in a short-beam bend test can be explained in terms of shear at the interface between fibre and resin. Attempts to obtain satisfactory compaction using plasma-treated HMPE fibre have not been successful. This is explained by the existence of a thin cross-linked surface layer on each filament that is produced by plasma treatment [5] which does not soften sufficiently when used in the compaction process and therefore does not produce high adhesion.

As the fibre content is reduced, the resin supports an increasing proportion of the shear strain and the ILSS approaches the value that would be expected for shear failure of the resin as indicated by the dotted lines in Figs 1–4. However, at the lower fibre levels, the tensile strain in the resin at the convex surface of the composite will exceed the resin tensile failure strain, and

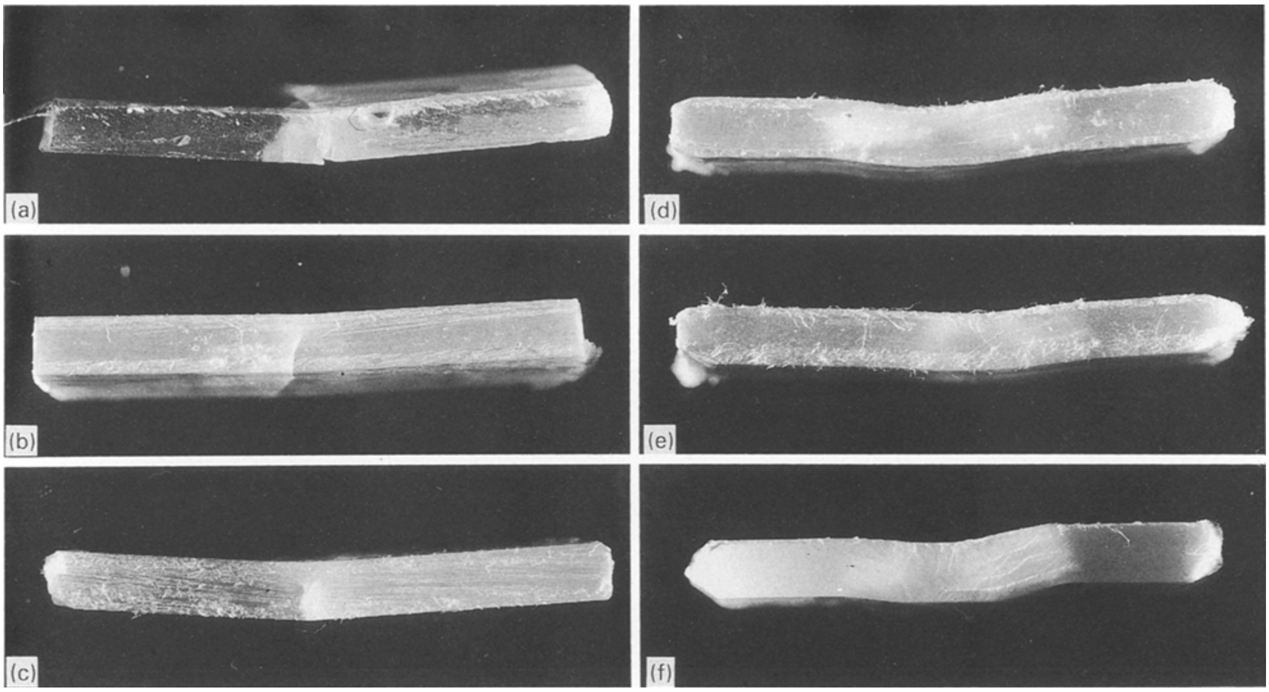

Figure 7 Photographs showing the effect of fibre volume fraction and plasma treatment on the type of failure for a Tenfor fibre composite; fibre volume fraction/ILSS: brittle failure, (a) no fibre, 17.0 MPa, (b) 0.18 untreated fibre, 31.7 MPa, (c) 0.18 plasma-treated fibre, 33.0 MPa; shear failure, (d) 0.45 untreated fibre, 18.4 MPa, (e) 0.45 plasma-treated fibre, 28.1 MPa, (f) 0.92 untreated compacted fibre, 17.2 MPa.

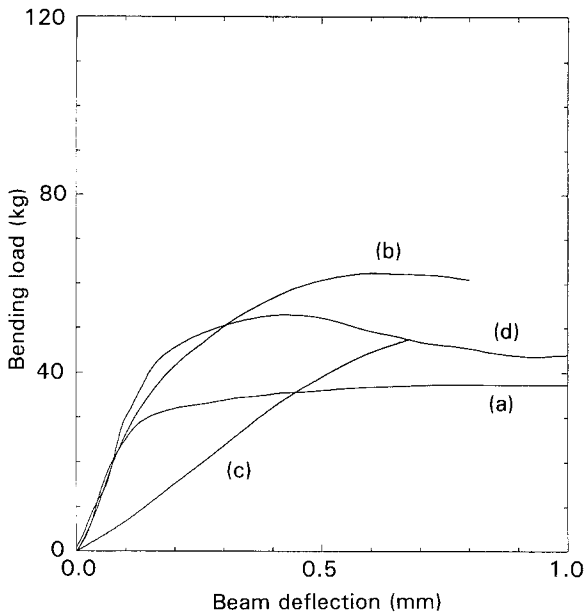

Figure 8 Load/deflection diagrams for a Tenfor fibre composite: (a) untreated 0.54 fibre volume fraction, (b) plasma-treated fibre, 0.54 volume fraction, (c) resin only, (d) 100% compacted fibre.

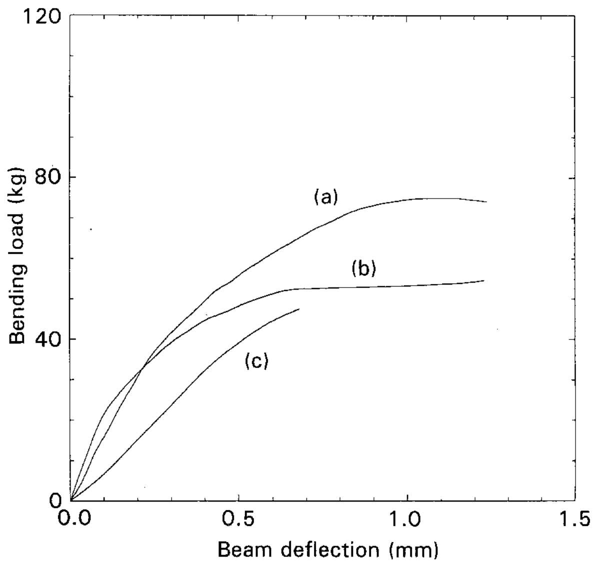

Figure 9 Effect of filament diameter on load/deflection diagrams for plasma treated fibre: (a) 0.015 mm diameter multifilament, 0.54 fibre volume fraction, (b) 0.055 mm diameter monofilament, 0.54 fibre volume fraction, (c) resin only.

brittle tensile failure of the resin will occur before the resin shear failure strength is reached. Thus, although the data in Figs 1–4 appear to extrapolate to values in the region of 59 MPa at zero fibre fraction, none of the results obtained exceed 40 MPa, and decrease towards the resin brittle failure stress at low fibre volume fractions.

Further understanding of the failure mechanism is obtained from observation of the short-beam bending curves. In Fig. 8, the bending load curves for untreated and plasma-treated Tenfor fibre are compared together with a resin only curve. The resin sample (curve c) gives an approximately linear curve terminating in brittle fracture at 0.7 mm deflection and a bending load corresponding to 17 MPa failure stress. With an untreated fibre composite (curve a), the bending load increases more rapidly as a result of the fibre reinforcement and has a marked shoulder before failing at a lower load than the resin sample. The plasma-treated sample (curve b) follows the path of the untreated sample (curve a) at low deflection as a consequence of the similar initial moduli of the fibre and resin components. However, the load then increases further and the sample finally fails at a similar deflection but greatly increased bending load. Observation of the samples during bending shows that no shear occurs before the shoulder of the curve and that beyond this point, shear increases rapidly until the peak load is reached and shear failure occurs. Thus, the untreated sample exhibits shear at 0.1 mm deflection whereas in the plasma-treated sample, shear is not apparent until the deflection is 0.5 mm, the higher deflection and load reflecting the increase in adhesion between fibre and resin produced by plasma treatment of the fibre surface.

The effect of increasing the filament diameter at constant fibre volume fraction is shown in Fig. 9. Increasing the filament diameter to 0.055 mm at constant fibre volume fraction will decrease the fibre surface area to approximately one-third of the 15 μm diameter filament surface area. This will greatly increase the filament surface stress and less deflection of the sample will be required to obtain fibre shear failure as seen in curve b.

Typical results for the overall effect of fibre volume fraction on the bending load curves for Tenfor and

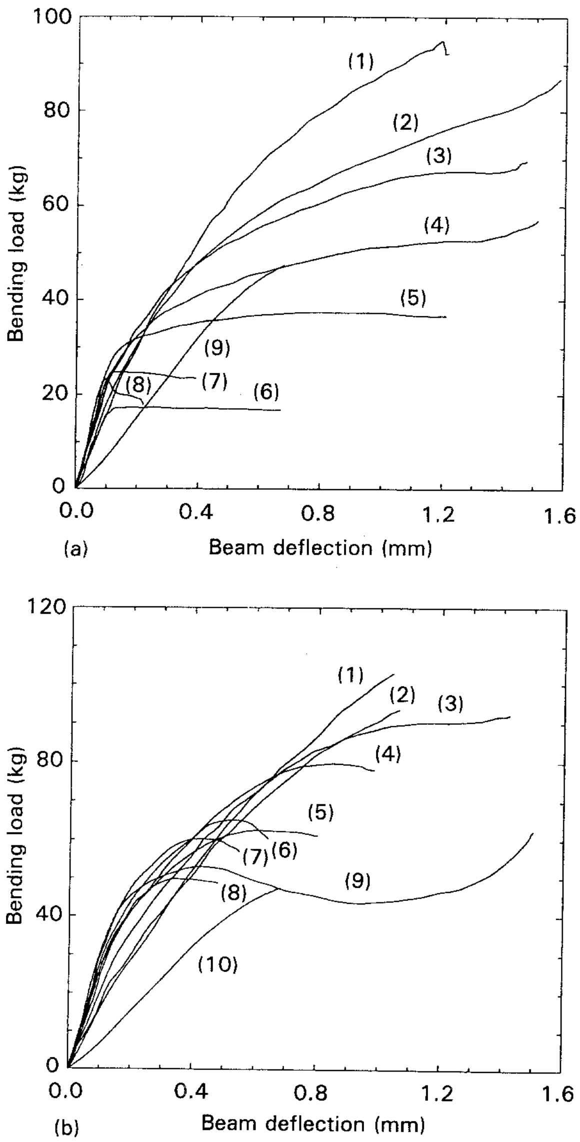

Figure 10 Load/deflection curves for Tenfor composites: (a) untreated, (b) plasma-treated. Fibre volume fraction: (1) 0.18, (2) 0.27, (3) 0.36, (4) 0.45, (5) 0.54, (6) 0.63, (7) 0.72, (8) 0.81, (9) 0.92 (compacted fibre), (10) no fibre.

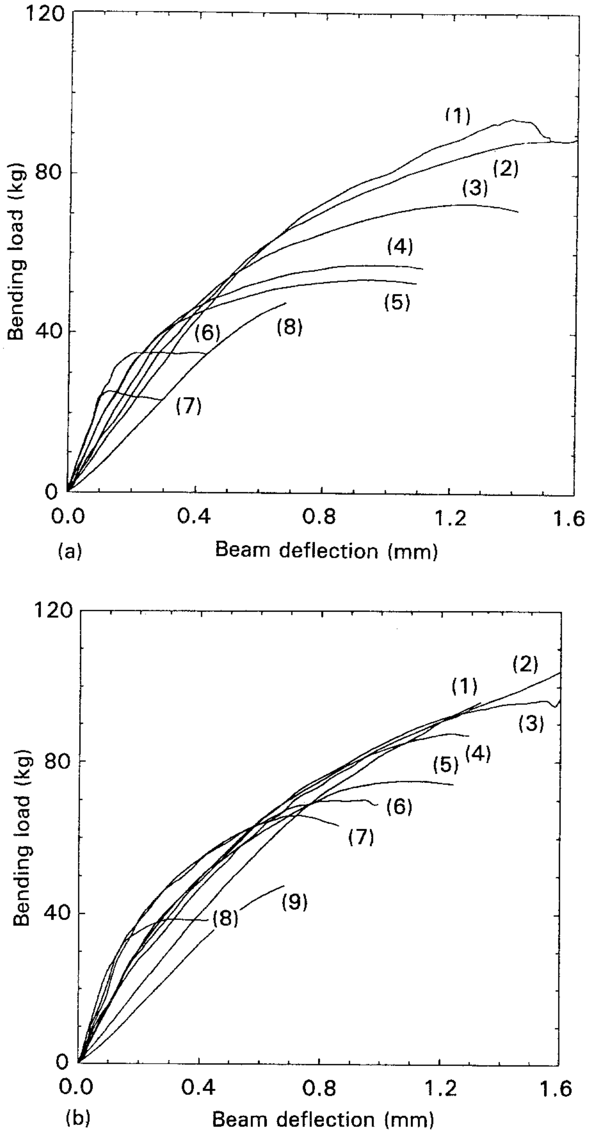

Figure 11 Load/deflection curves for multifilament composites: (a) untreated, (b) plasma-treated. Fibre volume fraction: (1) 0.18, (2) 0.27, (3) 0.36, (4) 0.45, (5) 0.54, (6) 0.63, (7) 0.72, (8) 0.92 (compacted fibre), (9) no fibre.

Multifilament fibres are shown in Figs 10 and 11. For both untreated fibres, it is seen that there is, in general, a broad shoulder on the curves and that the increase in bending load beyond this point where shear occurs is fairly gradual, reflecting the relatively poor adhesion between untreated fibre and resin. In the case of the plasma-treated fibres, the curves all have a clearly defined shoulder which lies close to the peak failure load, reflecting the greatly increased adhesion of the plasma-treated fibre to resin.

5. Conclusions

It was found that the shear failure stress of HMPE fibres in a unidirectional composite with epoxy resin, as measured by the interlaminar shear strength obtained from a short-beam bend test, increases after plasma treatment of the fibre but decreases with increasing fibre content and increasing filament diameter.

It is concluded from these observations that for fibre volume fractions greater than 0.3, the shear failure is controlled by the adhesion between the fibre and the resin. At fibre volume fractions below 0.3, on the other hand, the failure is increasingly dominated by the shear failure of the resin, with the ILSS values extrapolating towards the higher resin shear failure stress. However, the correspondingly higher failure strain of the composite at low fibre volume fraction results in a high surface stress which exceeds the resin brittle failure stress and results in a lower failure stress for the composite.

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