

Determination of interfacial shear strength in fibre-reinforced thermoplastic composites

M. J. Folkes and W. K. Wong

Department of Materials Technology, Brunel University, Uxbridge, Middlesex, UK
(Received 14 July 1986; revised 19 November 1986; accepted 2 December 1986)

A test method, suitable for the routine assessment of fibre-matrix bond strength of thermoplastic composites, has been developed. The method is sensitive enough to detect changes in interfacial bond strength resulting from different levels of silane applied to the glass fibre and the formation of transcrystalline morphology around the fibres. In addition to the chemical and physical nature of the interface, it was found that the shear yield strength of the matrix, fibre diameter and conditions used in sample fabrication all have important roles in determining the interfacial bond strength. These findings cast doubts on the validity of methods for predicting the strength of short-fibre-reinforced composites.

(Keywords: interfacial shear strength; critical fibre length; thermoplastic composites; silane levels; interfacial morphology; fibre diameter)

INTRODUCTION

The fibre-matrix interfacial shear strength is a very important parameter in controlling the strength and toughness of a composite material. Its magnitude is particularly affected by the fibre surface treatment and by any special 'size' applied to the fibre during manufacture^{1,2}. Recently, Miwa *et al.*³ have reported on the important role played by the interfacial shear strength in determining the strength of a composite. Also, the work of DiBenedetto⁴ demonstrates, quite remarkably, the effect of different fibre surface treatments on the interfacial shear strength.

In the case when the matrix is a crystalline thermoplastic, the interfacial shear strength may be additionally influenced by the morphology of the matrix close to the fibre surface^{5,6}. Although the interfacial morphology for many fibre-matrix combinations has received considerable attention, the link between this and the mechanical properties of the interface has remained inconclusive.

Attempts have been made previously by other workers, such as Kelly⁷, Drzal *et al.*⁸, Arridge⁹, Broutman¹⁰ and Hughes¹¹, to measure directly the interfacial shear strength (or critical fibre length) between fibre and matrix. Most of these studies are based on the measurement of fibre pull-out lengths or the fibre fragment lengths of a stressed specimen, which consists of a single fibre embedded in a *thermoset resin*.

The fabrication of resin blocks embedded with a single fibre is very time-consuming and by no means easy to accomplish. In order to overcome this, we prepared our specimens, which are in the form of thin tapes with well aligned fibres sparingly dispersed in a *thermoplastic* matrix, by an actual extrusion process. This method of sample preparation is quick and easy to perform. Also, any effect that melt processing may have on the fibres or interface during sample preparation (such as creating flaws on the fibre surface or removal of the 'size' coating, etc.), will be simulated using this preparation method.

Accordingly in this paper, a programme of work is

described, in which we have endeavoured to determine accurately the critical fibre length of various samples of glass fibre in a polypropylene matrix. This was carried out in order to investigate the following factors, which may affect the interfacial shear strength: (1) matrix morphology around the fibre; (2) size formulation; (3) silane level applied to the fibre; (4) fibre diameter; and (5) matrix shear yield strength.

THEORY

Figure 1 shows schematically a single fibre embedded in a matrix. When an external stress is applied to the specimen parallel to the fibre axis, the tensile load will be transmitted from the matrix to the fibre through the interfacial shear stress. An increase in the applied stress will result in a progressive break-up of the fibres into shorter fragments and a knowledge of the distribution of fragment lengths enables a value for the critical fibre length and hence interfacial shear strength to be determined. On the basis of the Kelly-Tyson model⁷, the tensile stress (σ_x) in the fibre increases linearly from nearly zero at the fibre ends to a maximum value, limited by the ultimate tensile strength of the fibre (σ_f). When this limit is reached, the fibre should break anywhere between A and B. On the other hand, the interfacial shear stress (τ_u), which has a maximum value at the fibre ends, decreases to nearly zero towards the middle of the fibre. If the external stress is increased, this fibre breakage process should continue until all fibre fragments are less than $2X_0$, in which case the tensile stress in the fibres cannot reach σ_f . The minimum fibre length for which the tensile stress can reach the ultimate tensile strength of the fibre is termed the 'critical length', l_c .

The Kelly-Tyson model predicts that the fibre fracture process should result in a distribution of fragment lengths from $l_c/2$ to l_c . Often, the distribution of fragment lengths obtained is broader than the 2:1 ratio predicted. This has been attributed to the existence of flaws in the fibre, thereby causing its strength to depend on length¹².

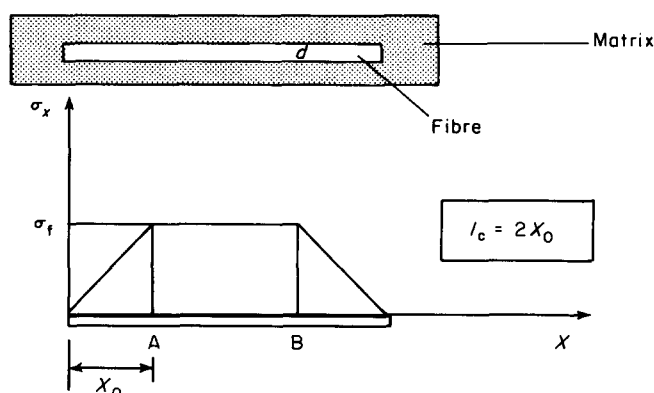


Figure 1 Schematic diagram showing the stress distribution along a fibre embedded in a matrix (load parallel to fibre axis)

Different models^{3,4} have been reported which take into account the existence of flaws along the fibre when utilizing the fibre fragment data for the evaluation of l_c . However, if a normal fibre fragment distribution histogram is obtained, the peak of the histogram should equal $\frac{3}{4}l_c$. Also, the midpoint (50% point) of the cumulative fibre fragment length distribution curve should also equal $\frac{3}{4}l_c$, if a large enough number of fragments are included.

Kelly and Tyson have related the critical length, l_c , to the interfacial shear strength through a force balance around the embedded fibre. They assumed that the shear stress at the interface (τ_u) is constant and equal to the shear yield strength of the matrix and obtained the following equation for critical length:

$$l_c = \sigma_f d / 2\tau_u$$

where d is the diameter of the fibre.

The Kelly-Tyson model seems to be of relevance to our system in determining interfacial bond strength from the fibre fragment lengths, since the thermoplastic matrix we use has a yield point.

The Kelly-Tyson model indicates that the shear yield strength of the matrix should be large enough to resist the shear stress at the interface, otherwise matrix yielding will occur before the maximum stress can be transferred to the fibre. In general, the shear stress at the interface can never exceed that of the shear yield strength of the matrix. When utilizing the Kelly-Tyson relationship, the ultimate tensile strength of the fibres can only be measured for pristine fibres. The true σ_f value of the fibres in a moulded component may be different.

EXPERIMENTAL

Materials

The samples of polypropylene used in this work were supplied by ICI and designated GW522M. Another special polypropylene powder premix designed for blending with silane-coated glass fibres to give 'coupled' reinforcement and a toughened-grade polypropylene with a lower shear yield strength were also used. The glass fibres (A, C, X, D and D_s) used are all of type E. Glass fibres A, C, D and D_s are all in a roving form. They were dry blended with the coupled-grade polypropylene for the preparation of the tape specimens. Fibre X was in the form of short composite rods which consisted of glass fibres wetted by some coupled grade polypropylene.

These rods were also dry blended with the GW522M polypropylene prior to the extrusion of the tape specimen. Some details of the materials are given in Tables 1 and 2.

Sample fabrication and testing

Fabrication of the specimens, which were in the form of a thin tape with fibres finely dispersed and well aligned, was accomplished using a Betol 2520EX extruder coupled with a slit die. Figure 2 shows the essential features of the extruder. Fibres were introduced together with the matrix into the Betol extruder through the feed hopper. The fibres were well separated and with a length at least five times the estimated critical fibre length. The temperature of the barrel was set at 175°C and that of the die zone adjusted to 250°C. The extruded polymer, in semi-molten form, was allowed to pass through a set of water-cooled nip rollers to be pressed into a thin tape. The speed of the nip roller could be varied to alter the thickness of the tape. However, care was taken not to stretch the tape too much during preparation. This could cause premature fibre breakage. During the extrusion process, most fibres were broken up into lengths of a few millimetres. To avoid severe fibre breakage, the extrusion speed was fixed at a low value. The action of the die and nip rollers tended to align the fibres in the direction of extrusion.

An area of the thin tape was selected and cut into a dumb-bell shaped specimen ready for straining, which was carried out using a Type W Monsanto Tensometer equipped with vice-type chucks for gripping films. During all tensile tests, care was taken to maintain a steady and uniform straining rate. Each specimen was subjected to an 8% tensile strain, which was greater than the fibre's ultimate tensile strain. The strain rate of each specimen was about 1 cm min⁻¹. After the test, the fibres in the specimen were broken into a large number of fragments.

Fibre fragment lengths were measured using an optical microscope fitted with a graticule eyepiece. Only fibres that were aligned fairly well along the tensile loading axis were selected for measurement. Also, short fibres which had only broken into two fragments were not included, as they may not be representative. An optical micrograph of a glass fibre in the polypropylene matrix after specimen pulling is shown in Figure 3. The breakage points can be distinguished easily as white spots along the fibre.

Table 1

PP sample	Tensile yield strength (MPa)	Shear yield strength (MPa)
GW522M	35	21
Special premix	35	21
Toughened grade	21	14

Table 2

Glass fibre	Silane level (%)	Average ultimate strength (GPa)	Diameter (μm)
A	—	3.4	13
C	—	3.4	13
X	—	3.4	10
D(1)	1	3.4	18
D(1.75)	1.75	3.4	18
D(2.5)	2.5	3.4	18
D _s (2.5)	2.5	3.4	9

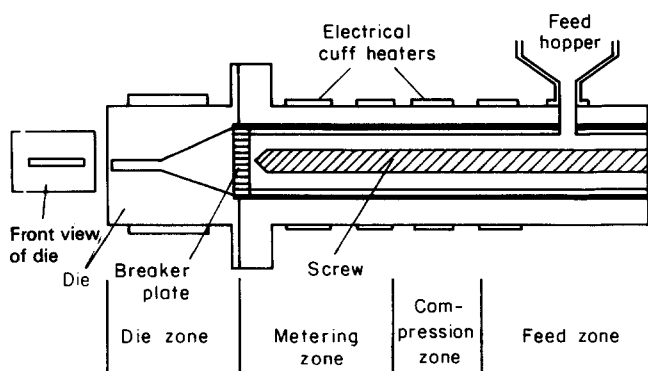


Figure 2 Schematic diagram showing the essential features of the single screw extruder and slit die

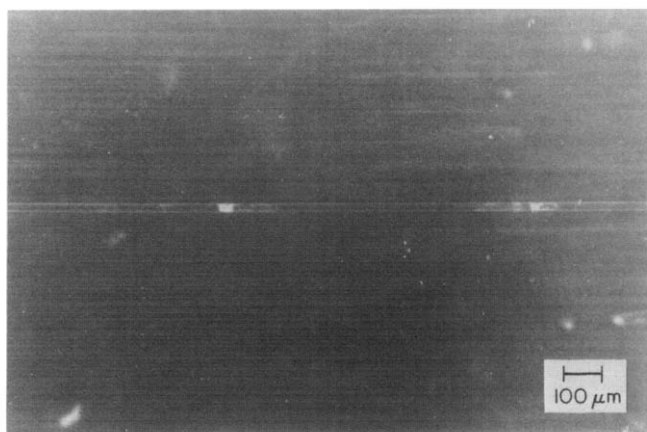


Figure 3 An optical micrograph of a glass fibre in a polypropylene matrix after specimen straining

RESULTS AND DISCUSSION

The fibre fragment length distribution histograms for the fibres, A, C, X, D and D_s using polypropylene as matrix are shown in Figures 4a, 5, 6, 7 and 8. The fibre fragment length corresponding to the peak of each histogram was multiplied by a factor of $\frac{4}{3}$ and recorded in Table 3 as l_{c1} . The fibre fragment length distributions for fibres A, C, D and D_s were measured from tapes prepared by using the *coupled-grade* polypropylene as matrix. For fibre X, tapes prepared by using the *non-coupled-grade* polypropylene were used. The fibre fragment length distribution for fibre A using tapes prepared from the *toughened-grade* polypropylene was also measured and the corresponding histogram is included in Figure 4a for comparison. The cumulative fibre fragment length distribution curve for fibre A is shown in Figure 4b. Similar curves for Figures 5–8 were also produced (not shown for purpose of conciseness). The points which represent 50% of the total number of fragments in each of these curves were multiplied by a factor of $\frac{4}{3}$ and recorded in Table 3 as l_{c2} . By comparing the values of l_{c1} and l_{c2} it was noted that, in the case when the histogram shows a normal distribution, these values agree very well. However, when the histogram shows some degree of skewness, a difference in l_{c1} and l_{c2} was always found. The skewness of these histograms, which is always towards the right, indicates the presence of a larger proportion of long fibre fragments. Hence, the skewness observed seems to have resulted from better fibre strength within the specimen.

The extruded specimen which had not been subjected to any further heat treatment did not reveal any specific morphology under an optical microscope. It was thought to consist of very small spherulites. Attempts were made to induce transcrystalline growth of the polypropylene matrix around the fibres A and C by heat treating the specimens produced. However, this was not successful due to the presence of coupling agent in the matrix, which can act as a nucleating agent and thus compete with the fibre surface for spherulitic growth. Thus, instead of transcrystalline morphology around the fibres, there appeared to be a large number of small spherulites around the fibres and throughout the matrix. Heat treatment of the glass fibre X-GW522M specimen to induce transcrystallinity proved to be successful because there is only coupled polymer on the fibre but not throughout the matrix.

Table 3 summarizes the critical lengths derived from the fibre fragment length measurements for each specimen.

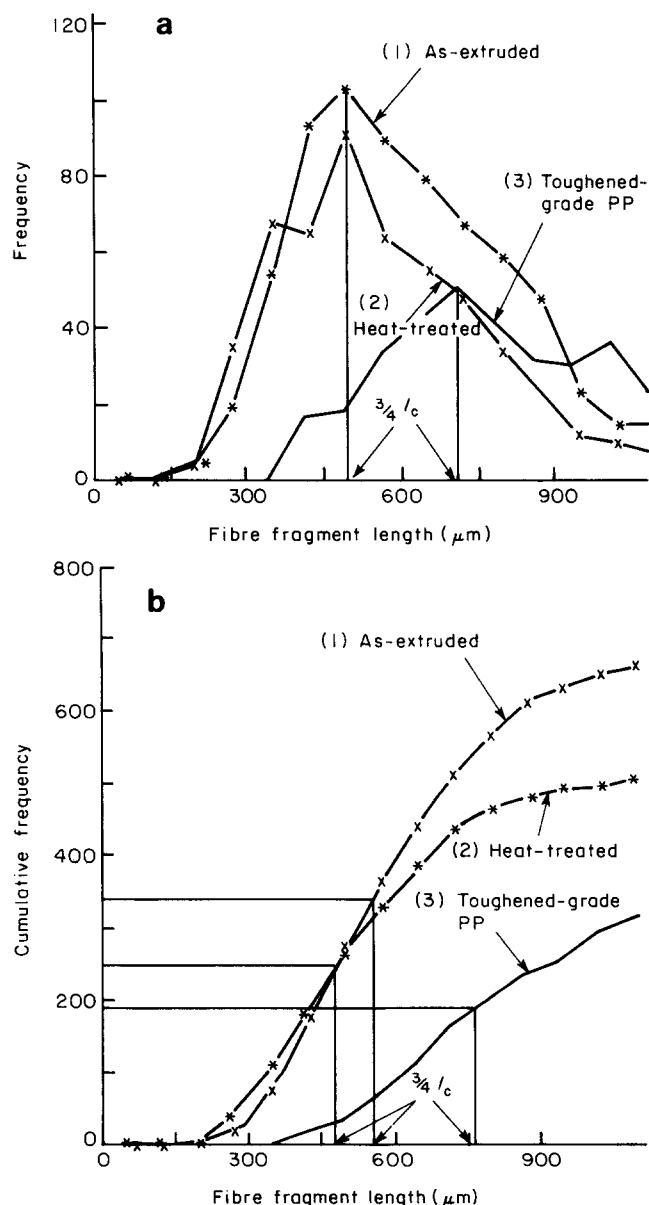


Figure 4 (a) Histogram of fibre fragment length distribution for fibre A in polypropylene. The numbers against the plots refer to the samples as defined in Table 3. (b) Cumulative fibre fragment length distribution curve for fibre A in polypropylene. The numbers against the plot refer to the samples as defined in Table 3

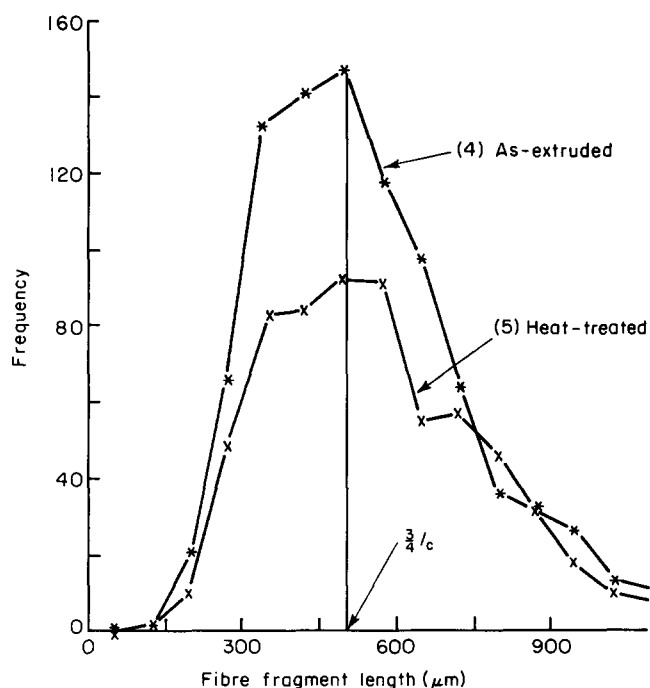


Figure 5 Fibre fragment length distribution for fibre C in polypropylene. Remaining details as for Figure 4

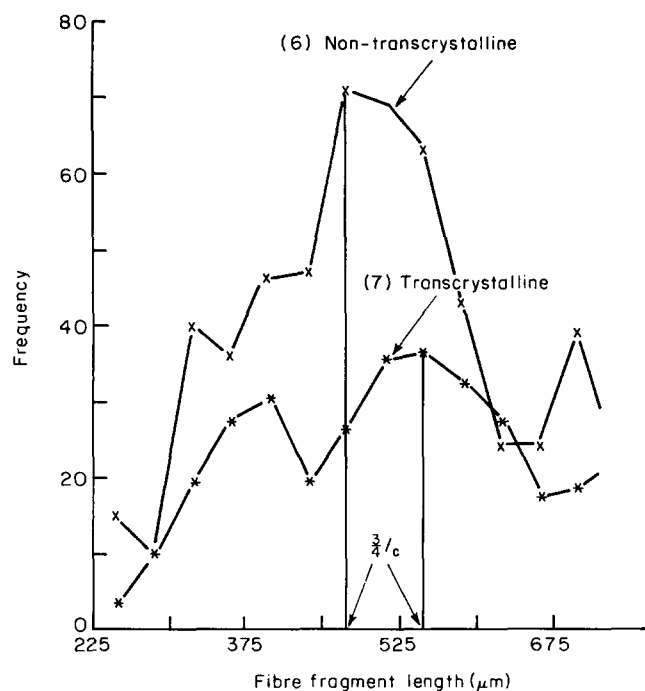


Figure 6 Fibre fragment length distribution for fibre X in polypropylene showing the effect of transcrystallinity on critical fibre length. Remaining details as for Figure 4

The values of critical length obtained from the peak of the histogram (l_{c1}) and the 50% point of the cumulative curve (l_{c2}) are shown. In column 6 of the table, the interfacial shear strength values (τ_u) are obtained from the measured critical fibre lengths (l_{c1}) by assuming the fibre strength to be that of the starting fibre before being subjected to the compounding process used in the tape manufacture. In column 7 of the table, the apparent fibre strength values (σ_f) in the tape samples were derived from the measured critical fibre lengths (l_{c2}) and by assuming that the maximum value of interfacial shear strength equals the shear yield strength of the matrix (l_{c2} was chosen for calculating the apparent fibre strength because this value

takes into account possible skewness in the fibre fragment histograms). It is rather surprising at first sight that the derived values of interfacial shear strengths shown in Table 3 are all higher than that of the shear yield strength of the polypropylene used. This anomaly, however, can be explained by taking into account the unrealistic assumption that the tensile strength of the glass fibre in the specimens is equal to that of the glass fibre in a pristine condition. The extrusion and subsequent nip-rolling action must have caused damage to the fibres and reduced their tensile strength. In fact, the apparent σ_f values derived from the l_{c2} values are much less than the tensile strength of the starting fibres. Fibre D_s has an apparent σ_f value of 3.4 GPa, which equals the tensile strength of a pristine fibre. The explanation of this result will be discussed later.

It is clear from the results obtained that samples 1, 4 and 6 all give the same l_{c1} value. These samples correspond to the as-extruded specimen of fibres A, C and X in coupled-grade polypropylene. The similarity of l_{c1} values for these samples indicates that the fibres A, C and X, although having a different size formulation on their

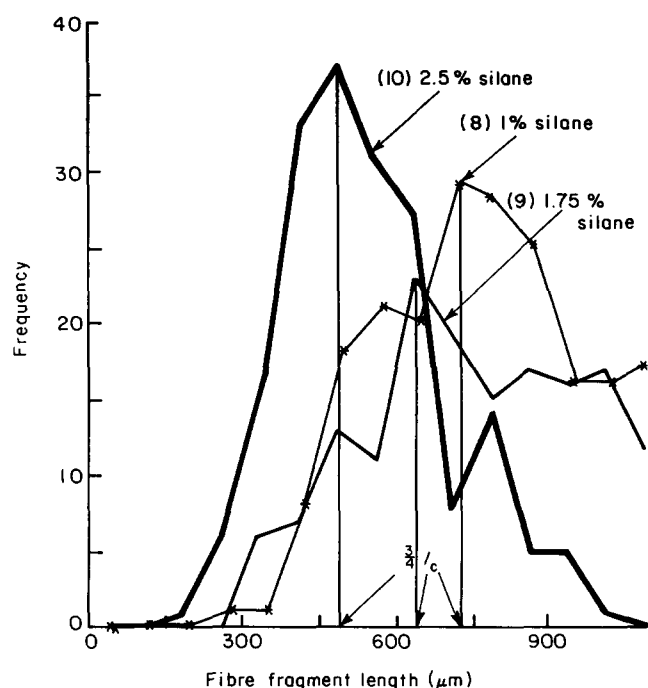


Figure 7 Fibre fragment length distribution for fibre D in polypropylene showing the effect of silane level on critical fibre length. Remaining details as for Figure 4

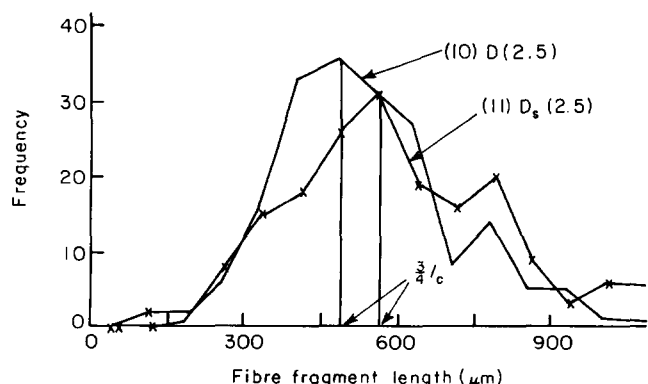


Figure 8 Fibre fragment length distribution for fibres D and D_s in polypropylene showing the effect of fibre diameter on critical fibre length. Remaining details as for Figure 4

Table 3

Sample	Heat treatment	Morphology around fibre	l_{c1}^a (μm)	l_{c2}^b (μm)	τ_u (MPa)	Apparent σ_f (GPa)
(1) Fibre A/coupled PP	No	Very small spherulites	650	720	34.0	2.3
(2) Fibre A/coupled PP	Yes	Small spherulite	650	640	34.0	2.1
(3) Fibre A/toughened PP	No	Very small spherulites	950	1010	23.3	2.2
(4) Fibre C/coupled PP	No	Very small spherulites	650	625	34.0	2.0
(5) Fibre C/coupled PP	Yes	Small spherulites	650	645	34.0	2.2
(6) Fibre X/non-coupled PP	No	Very small spherulites	650	650	26.2	2.7
(7) Fibre X/non-coupled PP	Yes	Transcrystalline	750	700	22.7	2.9
(8) Fibre D (1% silane)/coupled PP	No	Very small spherulites	950	1010	31.9	2.4
(9) Fibre D (1.75% silane)/coupled PP	No	Very small spherulites	860	1090	35.6	2.5
(10) Fibre D (2.5% silane)/coupled PP	No	Very small spherulites	660	670	46.4	1.6
(11) Fibre D _s (2.5% silane)/coupled PP	No	Very small spherulites	760	730	20.1	3.4

^aCritical length obtained from the peak of the histograms

^bCritical length obtained from the 50% point of the cumulative curves

surfaces, exhibit very similar interfacial shear strengths. Since the τ_u values derived for the three samples are all very high, it is probable that the interfacial bond strengths of these three fibres are all very large and exceed the shear yield strength of the polypropylene matrix. Hence, the similar l_{c1} values obtained merely reflect the shear yield strength of the matrix, which is the limiting factor controlling the maximum interfacial shear strength possible for any fibre-matrix composite. The above argument is confirmed by the result obtained for sample 3, in which a toughened-grade polypropylene of lower shear yield strength was used as the matrix. The increase in l_{c1} value for this sample when compared with sample 1 cannot be due to a reduction in interfacial bond strength, since the same fibre and coupling agent were used. Hence, the apparent reduction of interfacial shear strength in sample 3 must be caused by the lower shear yield strength of the matrix. This result has very important implications as it indicates that further improvements in interfacial shear strength between fibre and matrix will be limited by the shear yield strength of the matrix material used.

The results for samples 1, 2, 4 and 5 in Table 3 indicate that spherulite size around glass fibres has no significant effect on its critical length. However, the formation of transcrystallinity around the glass fibre seems to have affected the critical fibre length and hence the interfacial shear strength. The increase in critical fibre length for sample 7 may be even more significant if completely uniform transcrystallinity can be induced along the glass fibres. Owing to the intrinsic poor ability for glass fibre to nucleate columnar growth¹³, the transcrystalline morphology is only made possible by the presence of coupling agent, which is a good nucleating agent, on the surface of glass fibre X. Hence, the resulting transcrystallinity around the fibres was not apparent in some places even along the same glass fibre. From an examination under an optical microscope, there were some indications that fibre breakage tended to occur in places where transcrystallinity is absent. The mechanism by which transcrystalline morphology leads to a general degree of poor bonding may be similar to that reported by Bessel, Hull and Shortall¹⁴. They observed from their carbon fibre-nylon-6 composites a surface of weakness about 2 μm from the fibre surface, which was related to the transcrystallinity around the fibre. The fibres that had pulled out from the fracture surface were also observed to be surrounded by a thin sheath of nylon. It is thus possible that the increase in critical fibre length for the

transcrystalline specimen observed in our studies is due to the existence of a thin inner layer in the transcrystalline zone, which has a low adhesive strength to the outer layer of the transcrystalline material.

The difference in l_{c1} values obtained for fibres D(1), D(1.75) and D(2.5) shows the effect of silane concentration on the interfacial bond strength between the fibre and the matrix. The interfacial bond strength increases with the silane level applied.

According to the Kelly-Tyson equation, a reduction in fibre diameter will result in a proportional decrease in critical fibre length, if τ_u remains the same. However, from the experimental results obtained, such a relationship was not found (compare samples 10 and 11). This deviation from the Kelly-Tyson equation must be due to the effect of fibre diameter on the apparent fibre tensile strength. A small-diameter fibre should, statistically speaking, have fewer flaws than a larger fibre. However, most important of all, a small flaw size, which causes a reduction in tensile strength in a large-diameter fibre, may result in total fibre failure in a smaller-diameter fibre when subjected to tensile stress. During the preparation of the specimens, a certain amount of tensile stress will be applied to the tape, which is unavoidable. If flaws that can cause total fibre failure in the small-diameter fibre (9 μm) do occur along this fibre, we may expect the extruded tape to consist of shorter fibres (in comparison with 18 μm fibre). This was indeed found to be the case. Furthermore, these short fibre fragments of the small-diameter fibre should be relatively flawless (most flaws that originally existed along the fibre surface led to the fibre breakage). This argument is supported by the high apparent σ_f value obtained for the small-diameter fibre.

CONCLUSIONS

A test method has been developed which enables the rapid determination of critical length of glass fibres in a polypropylene matrix. The method is sensitive enough to detect changes in interfacial bond strength which result from different levels of silane applied on glass fibres. Spherulite size around glass fibres has no significant effect on the critical fibre length. However, transcrystalline morphology induced around the fibres causes a reduction in interfacial bond strength. The different size formulations used in this work do not seem to promote different interfacial bond strengths. This is due to the fact that all the size formulations used give

interfacial bond strengths greater than the shear yield strength of the polypropylene matrix. A large reduction in interfacial shear strength is shown between fibre A and the toughened-grade polypropylene, which has a lower shear yield strength. This indicates that the interfacial shear strength is limited by the shear yield strength of the matrix. Hence, further improvement in the fibre-matrix interfacial bond strength will be limited by the matrix material used. In all the tests carried out, the interfacial shear strengths derived are higher than the shear yield strengths of the matrix. These unrealistically high values are due to the reduction of fibre tensile strength during the extrusion process. In the case of small-diameter fibres, flaws inflicted on the fibre during processing caused total fibre failure to occur. The reduction of fibre tensile strength during processing casts doubts on the validity of methods for predicting the strength of short-fibre-reinforced composites.

In summary, the method has proven to be highly accurate and is suitable for the routine assessment of the fibre-matrix bond strength. We believe that the technique is more generally applicable to other fibre-thermoplastic resin combinations.

ACKNOWLEDGEMENTS

We would like to express our sincere thanks to SERC for financial support, without which this work could not have been carried out. Also, we extend our thanks to ICI and

Fibreglass Ltd for supplying us with materials. Last but not least, we thank Mr D. W. Grinter for carrying out part of the experimental work.

REFERENCES

- 1 Morrell, S. H. *Plast. Rubber Process. Appl.* 1981, 1, 179
- 2 Shau-Zon Lu, 35th Annu. Tech. Conf. Reinforced Plastic/Composites Institute', The Society of the Plastics Industry, Inc., 1980
- 3 Miwa, M., Ohsawa, T. and Tahara, K. *J. Appl. Polym. Sci.* 1980, 25, 795
- 4 DiBenedetto, A. T. Paper presented at the '28th Microsymp. on Macromolecules-Polymer Composites', Prague, 1985
- 5 Hudson, M. G. and McGill, W. J. *J. Polym. Sci., Polym. Phys. Edn.* 1985, 23, 121
- 6 Campbell, D. and Qayyum, M. M. *J. Mater. Sci.* 1977, 12, 2427
- 7 Kelly, A. and Tyson, W. R. *J. Mech. Phys. Solids* 1965, 13, 329
- 8 Drzal, L. T., Rich, M. J., Camping, J. D. and Park, W. J. '35th Annu. Tech. Conf. Reinforced Plastics/Composites Institute', The Society of the Plastics Industry, Inc., 1980
- 9 Takaku, A. and Arridge, R. G. C. *J. Phys. D. Appl. Phys.* 1973, 6, 2038
- 10 Broutman, L. J. and Krock, R. H. (Eds.) 'Modern Composite Materials', Addison-Wesley, New York, 1967
- 11 Hughes, J. D. H. Applied Chem. Division, AERE, Harwell, 1977, Report No. R8683
- 12 Fraser, W. A., Ancker, F. H., DiBenedetto, A. T. and Elbirli, B. *Polym. Compos.* 1983, 4, 238
- 13 Burton, R. H. and Folkes, M. J. *Plast. Rubber Process. Appl.* 1983, 3, 129
- 14 Bessell, T., Hull, D. and Shortall, J. B. *Faraday Spec. Disc., Chem. Soc.* 1972, 2, 137