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# The mechanical properties of glass/polypropylene multilayer laminates

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Multilayer laminates have been fabricated from thin sheets of glass and polypropylene. By the use of a special surface treatment applied to the glass surfaces, fully transcrystalline polypropylene layers could be formed. This facilitated the interpretation of the mechanical properties of the laminates in terms of the microstructure of the polymeric layers. It was found that the presence of transcrystalline layers leads to modest increases in flexural modulus and strength, and strain energy release rate $G_c$. These laminates can be valuable models for the mechanical properties of fibre-reinforced thermoplastics.

## 1. Introduction

The use of fibre-reinforced materials to produce strong components from carbon, glass and organic fibres and polymer matrices is now well established. Although such composites are being used successfully, their full potential has yet to be realized.

The mechanical properties of fibre-reinforced materials crucially depend on the fibre length distribution, fibre orientation distribution and the fibre/matrix interfacial shear strength. A common assumption is that the polymer matrix is largely unaffected by the presence of the fibres. This is certainly not true in the case of thermoplastic polymers, because microstructural modifications can occur close to the fibres. A fibre embedded in a thermoplastic melt can act as a nucleant for the growth of spherulites. If there are many nucleation sites along a fibre's surface, the resulting spherulite growth will be restricted in the lateral direction, so that a columnar layer, known as transcrystallinity [1] will develop and enclose the fibre.

Some workers [1–5] have systematically studied the growth of transcrystallinity around various fibres. In general, carbon and organic fibres readily nucleate transcrystallinity, while glass fibres give rise to it only occasionally. Usually where a glass fibre has been moved in relation to the melt, transcrystallinity has been observed after crystallization has taken place [6, 7]. Campbell and White [8] suggest that these observations are the result of the orientation of molecular chains, due to localized flow occurring at the fibre–melt interface.

Bessell and co-workers [2, 3] have studied the effects of transcrystallinity on the performance of glass and carbon-fibre-filled nylon 6. After tensile testing, they observed that on the fracture surfaces of the specimens, there were 2 $\mu$m thick layers of nylon 6 around those types of fibre where transcrystallinity was present. It appears that transcrystalline layers may possess areas of weakness between an inner and an outer zone. Folkes *et al.* [9] may well have observed the same phenomenon in carbon-fibre-filled nylon 6,6 following short beam shear tests.

Some studies have also been made of the effects of transcrystallinity on polymer/substrate bond strength. It has been found with carbon-fibre-filled nylon 6,6 [9] and glass-fibre-filled polypropylene [10] that the fibre/matrix bond strength is reduced after the generation of transcrystallinity around the fibres. However, Masouka [11] with steel blocks bound with nylon 12, and Ritchie and Cherry [12] with glass blocks bound with high density polyethylene, have found that the generation (or otherwise) of transcrystalline layers makes little difference to bond strength.

It has also been reported that transcrystalline layers may form protective sheaths around fibres. In nylon- and polyester-fibre-filled polypropylene composites [13, 14] it has been reported that transcrystalline layers may prevent the deformation of fibres prior to failure. Huson and McGill [15], in a study of single copper fibres embedded in polypropylene, found that transcrystalline layers also seemed to prevent the deformation of fibres prior to pull-out. Although the fracture processes have been discussed where transcrystallinity is present, there has not been any quantitative evaluation of the fracture mechanics involved.

The reported protective mechanisms [13–15] may be a direct effect of the mechanical properties of the transcrystalline layers [16]. However, poor adhesion between fibres and matrices would also prevent the deformation of the fibres, as it is necessary for forces to be transferred from the matrix via an effective bond for the fibres to be broken or deformed.

Schonhorn *et al.* [17] have speculated that if the transcrystalline layers can be shown to be relatively strong, then they might be an advantage in obtaining good adhesion. In fact, the mechanical properties of transcrystalline layers, in the form of separate sheets, have been measured [16]. Transcrystalline layers tend to be highly crystalline and orientated, and as a result have a relatively high Young's modulus, shear and tensile yield strength but low elongation to break and low failure energy. This indicates that Schonhorn's speculation may turn out to be valid.

In view of our previous work on the characteriz-

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ation of the mechanical properties of transcrystalline films, it is of interest to establish how they might contribute to the overall performance of a composite. For this purpose, laminate composites can be constructed with polymer layers of accurately controlled thickness and morphology. Such model composites overcome the principal difficulty of producing predetermined morphologies in fibre-reinforced composites for which there is little control over fibre spacing. Accordingly, in this paper we report on the fabrication and testing of glass/polypropylene laminates which serve as convenient two-dimension analogues of glass-fibre-reinforced polypropylene.

## 2. Preparation of the laminate composites

### 2.1. Materials

The glass layers used in our composites were cover slips 100 to 130 $\mu$m thick. Their areal dimensions were 66 × 22 mm.

The polypropylene consisted of two grades, both supplied by ICI; a coupled grade used for enhancing the nucleation on the glass surfaces, and an uncoupled grade GW522M, used for the production of the layers. A silane coupling agent Z6032, supplied by Dow Corning Corporation, was used to enhance the bond strength between the glass and polypropylene layers.

### 2.2. Construction

Since our purpose was to compare the mechanical properties of glass/polypropylene laminates exhibiting transcrystalline and non-transcrystalline morphologies, a special procedure had to be devised whereby a transcrystalline morphology could be developed against a glass surface. Glass is generally regarded as a poor nucleating surface and so it was necessary for us to use a novel approach.

The glass sheets were treated with the silane coupling agent Z6032, which has been previously shown [18] to be suitable for promoting adhesion between glass and polypropylene. The glass sheets were first thoroughly cleaned with a detergent and then washed successively with distilled water, acetone and chloroform. The sheets were then dipped into a 1% solution of Z6032 and methanol. The methanol was then allowed to evaporate off.

The silanated glass sheets were then covered with a thin layer of coupled-grade polypropylene, for two reasons: first, to ensure a strong interfacial bond and second, to provide strong nucleation [10] at the interface in order to promote a transcrystalline morphology. The treatment consisted of dipping the silanated glass sheets into a 1% solution of the coupled grade of polypropylene in boiling decalin. The decalin was then allowed to evaporate off.

A tape of uncoupled-grade polypropylene, GW522M, having a thickness of 100 to 150 $\mu$m, was extruded using a Plasticizer Ltd Fibrillator. Laminates were constructed by stacking alternate sheets of treated glass and polypropylene, and holding them in compression between two sheets of Melinex and then two thicker pieces of glass.

Laminates of three layers of glass and four layers of polypropylene (3G4P), six layers of glass and seven layers of polypropylene (6G7P), and nine layers of glass and ten layers of polypropylene (9G10P), were constructed for the purpose of flexural testing. Some 6G7P laminates were also constructed for impact testing. For the determination of interlaminar shear strength, laminates of 14 layers of glass and 15 layers of polypropylene (14G15P) were constructed. In all cases there were polypropylene layers on the outside faces of the laminates.

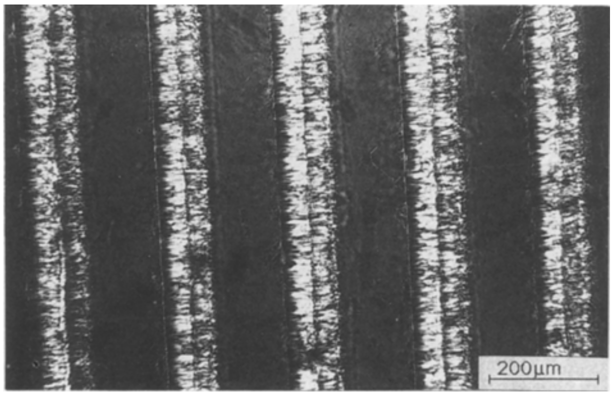

*Figure 1* Polarized light micrograph of a cross section through a transcrystalline polypropylene/glass laminate.

The laminates were then placed in an oven held at 200° C for 30 min in order to melt the polypropylene and to achieve a good interfacial bond. In order to generate transcrystallinity, they were then transferred to a separate oven held at 120° C for a further 10 min, after which they were briefly quenched in cold water. For comparison purposes, some fine spherulitic laminates were prepared by immediately quenching in cold water, after melting the polypropylene.

### 2.3. Microscopy

It was necessary to confirm the presence of the two microstructures in the laminates that had been prepared. Accordingly, some of the laminates were cut into thin slices (about 50 $\mu$m thick) using an Isomet diamond wafering saw. These were then examined edge on under crossed polars using a Reichert Zetopan microscope.

Fig. 1 shows a transcrystalline morphology where two transcrystalline zones have grown from the treated glass surfaces to meet in the middle of the polypropylene layer. Fig. 2 shows a fine spherulitic morphology throughout the sheets that have been quenched.

## 3. Mechanical testing of laminate composites

### 3.1. Flexural testing

3G4P, 6G7P and 9G10P laminates with transcrystalline and fine spherulitic morphologies were used for these tests.

During three-point-bending tests, two types of stresses occur in test specimens: tensile and compressive stresses ($\sigma$) on the outer faces and shear stresses ($\tau$) in the mid plane. The ratio of $\sigma/\tau$ depends on the test geometry according to:

$$\sigma/\tau = 2S/d \qquad (1)$$

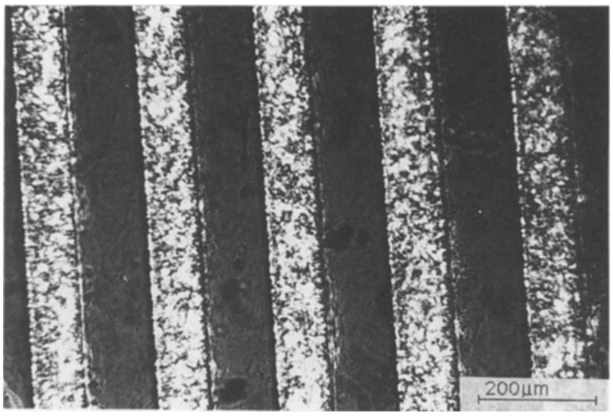

*Figure 2* Polarized light micrograph of a cross section through a fine spherulitic polypropylene/glass laminate.

where $d$ is the beam thickness and $S$ is the span. It follows that the likelihood of tensile failure will be increased by having a thin specimen or a large span during testing.

The span-to-depth ratios were to an extent controlled by the size of the laminates, but the 6G7P specimens in the test will have a span-to-depth ratio of about 32 : 1, which is the same as that recommended by ASTM D790, as used for the testing of fibre-reinforced plastics. The 3G4P specimens with a higher span-to-depth ratio are likely to display mainly tensile failure, while the 9G10P specimens would probably show this to a lesser extent.

The laminates were subjected to slow three-point-bending tests using the span-to-depth ratios shown in Table I. These tests were carried out on an Instron 1195 tensometer using a crosshead speed of 2 mm $\text{min}^{-1}$.

The flexural modulus, $E$, was calculated using the equation:

$$E = S^3m/4bd^3 \qquad (2)$$

where $b$ is the beam width and $m$ is the value of the applied load divided by the crosshead movement of the tensometer.

The flexural strength, $\sigma$, was calculated using the equation:

$$\sigma = 3PS/2bd^2 \qquad (3)$$

where $P$ is the maximum load applied.

### 3.2. Short beam shear testing

For the determination of interlaminar shear strength, which gives a measure of the degree of glass/polypropylene adhesion, 14G15P laminates having either transcrystalline or fine spherulitic morphology were used. These specimens had a span-to-depth ratio of 5 : 1 and this ensured mainly shear failure during testing.

The laminates were tested on a Monsanto 500 tensometer, using a crosshead speed of 5 mm $\text{min}^{-1}$.

The interlaminar shear strength, $\tau$, is given by

$$\tau = 3P/4bd \qquad (4)$$

### 3.3. Impact testing

For the measurement of impact strength, 6G7P laminates having the two different polypropylene microstructures were used. Tests were carried out on unnotched specimens using a Daventest instrumented drop-weight impact tester with a Charpy configuration, employing a 27 kg weight and a 2 m $\text{sec}^{-1}$ impact velocity. The values of impact strength, $I$, were determined using the equation

$$I = I_0/bd \qquad (5)$$

where $I_0$ is the energy required to break a laminate as recorded from the impact tester.

## 4. Discussion of results

### 4.1. Flexural testing

From Table I it can be seen how the microstructure of the polypropylene affects the flexural moduli and strengths of the laminate composites. The main differences appeared between the two types of morphology for the 3G4P and 6G7P laminates, though there was little difference in the flexural properties between the two types of 9G10P laminate. The laminates with transcrystalline polypropylene layers displayed only marginally higher flexural moduli than those with fine spherulitic polypropylene layers. However, the flexural strengths were significantly higher in the case of the laminates with transcrystalline morphologies; this being consistent with the tensile and shear properties of the two types of polypropylene layers described previously [16]. It would appear that the generation of transcrystalline layers does contribute an improvement to composite properties in terms of flexural strength.

A particularly interesting observation was made during the testing of the laminates. In both types of 3G4P and 6G7P laminates, evidence of multiple peaks on the stress/strain curves could be observed. A

TABLE I Mechanical properties of polypropylene/glass laminate composites, after three-point-bending tests

| Property | Sample | No of tests | Span to depth ratio | Type of polypropylene | |
|---|---|---|---|---|---|
| | | | | Transcrystalline | Fine spherulitic |
| Flexural modulus (GPa) | 3G4P | 6 | 60 : 1 | 20.9 | 20.4 |
| | 6G7P | 9 | 32 : 1 | 23.4 | 22.2 |
| | 9G10P | 6 | 22 : 1 | 26.3 | 26.3 |
| Flexural strength (MPa) | 3G4P | 6 | 60 : 1 | 100.2 | 84.6 |
| | 6G7P | 9 | 32 : 1 | 95.4 | 88.3 |
| | 9G10P | 6 | 22 : 1 | 87.1 | 88.7 |
| Interlaminar shear strength (MPa) | 14G15P | 6 | 5 : 1 | 6.5 | 5.4 |
| Impact strength (kJ $\text{m}^{-2}$) | 6G7P | 6 | 27 : 1 | 2.3 | 2.6 |

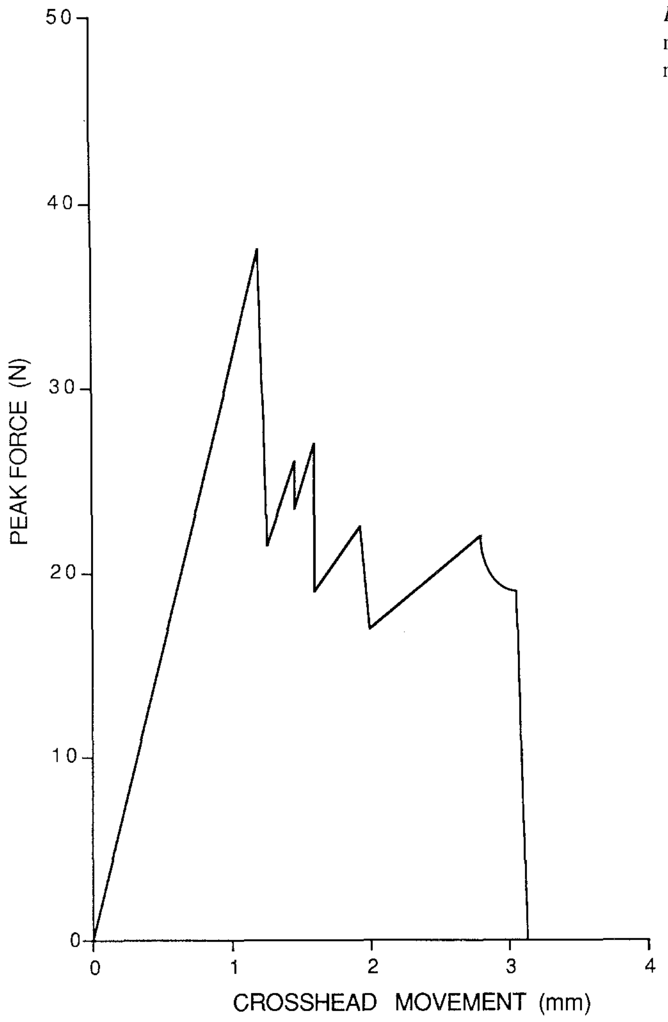

*Figure 3* Typical three-point-bending failure curve for a 6G7P laminate (with transcrystalline polypropylene). Note the presence of multiple failure peaks.

typical example of such a curve is shown in Fig. 3. With the 3G4P laminates, three peaks were often observed, which corresponded to the number of glass layers present. Similarly with the 6G7P laminates, six peaks were usually observed. With the 9G10P laminates it was difficult to attribute peaks to the number of glass layers present. It is apparent with the 3G4P and 6G7P laminates that one is seeing the sequential failure of glass layers. The same situation undoubtedly occurs with fibre reinforced polymers, except that there are too many fibres present to be able to detect individual breakages.

### *4.1.1. Fracture mechanics*

The sequential failure of the glass layers can be used as a basis for the evaluation of the fracture properties of the laminates, albeit under comparatively slow loading conditions. For each type of 3G4P and 6G7P laminate, the force values of the individual peaks ($P$) were plotted against the square of the effective thickness ($d^2$), where one can observe a linear relationship. The effective thickness is defined as the thickness of the unbroken part of the laminate prior to the breakage of a given layer. Such plots are shown in Figs 4–7. This linear relationship can be used to evaluate the critical strain energy release rate $G_c$ (see Table II).

The value of $G_c$ can be determined by using the equation below [19]

$$G_c = 9S^2 a\pi(1 - v^2)P^2/4d^4b^2E \qquad (6)$$

where $a$ is the crack depth, $v$ is the Poisson's ratio and $E$ is the Young's modulus of the laminate.

The values of the slopes, $C$ (see Table II) obtained from Figs 4–7, where $C = P/d^2$, may be substituted into Equation 6 to obtain $G_c$

$$G_c = 9S^2 a\pi(1 - v^2)C^2/4b^2E \qquad (7)$$

Since the specimens were unnotched, we have taken $a$ as being that corresponding to the intrinsic defect size in glass, i.e. about 10 $\mu$m. Other assumptions were also made regarding some of the constants in Equation 7, namely that the value of Poisson's ratio was assumed to be 0.3 and that the moduli values used were those determined experimentally in the flexure tests using Equation 2.

Previously only qualitative observations [13–15] have been made on the role of transcrystalline layers in toughening composites. It has been speculated, however, that transcrystalline layers may provide a

TABLE II Fracture properties of polypropylene/glass laminate composites, derived from slow three-point bending and impact tests

| Type of laminate | Slow three-point bending | | Impact tests |
|---|---|---|---|
| | $C$ (MPa) | $G_c$ (kJ m$^{-2}$) | $G_c$ (kJ m$^{-2}$) |
| 3G4P Transcrystalline | 16.2 | 4.16 | – |
| 3G4P Fine spherulitic | 12.0 | 2.34 | – |
| 6G7P Transcrystalline | 14.7 | 3.07 | 0.033 |
| 6G7P Fine spherulitic | 13.7 | 2.81 | 0.037 |

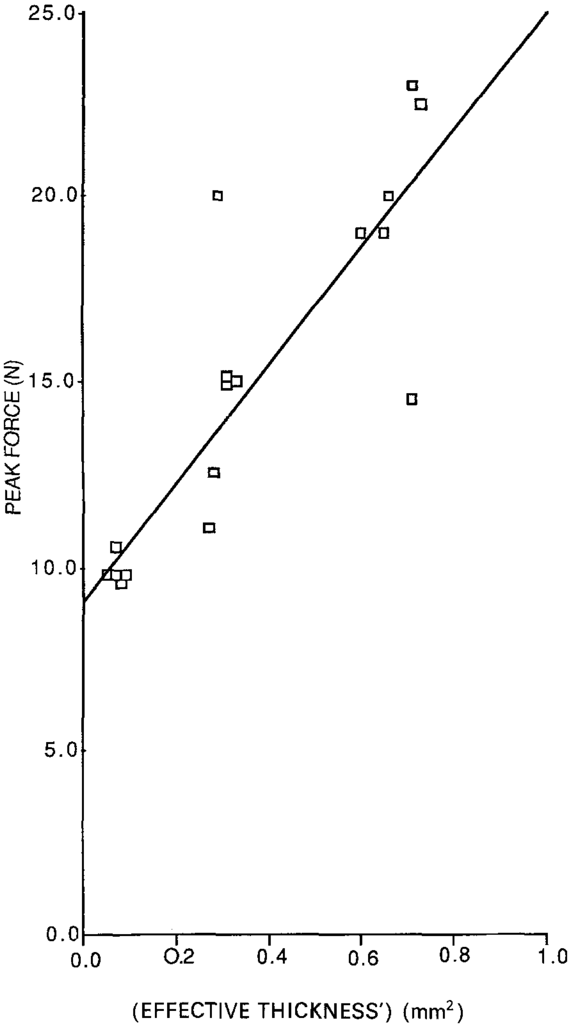

*Figure 4* Plot of failure peak force against the square of the effective thickness, for the determination of $G_c$ for a 3G4P laminate (with trancrystalline polypropylene).

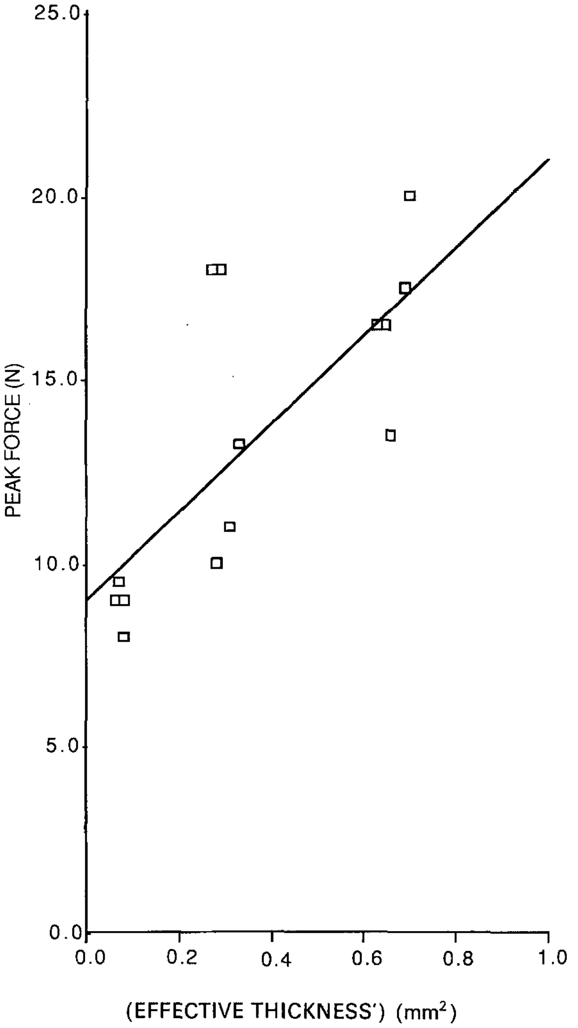

*Figure 5* Plot of failure peak force against the square of the effective thickness, for the determination of $G_c$ for a 3G4P laminate (with fine spherulitic polypropylene).

crack-stopping mechanism [14, 20, 21] which could contribute to this toughening. For the first time, it is apparent from our quantitative analysis (see Table II) that the generation of transcrystalline layers can increase the value of $G_c$ of a composite. It has been found previously [16] that transcrystalline layers require relatively little energy to fail. This is not consistent with the findings here, although intuitively one may expect some differences as the test methods employed were quite different.

While it is true that transcrystalline layers crack easily [16], in the situation of a composite undergoing three point bending they may experience multiple cracking, in the form of many new cracks or microcracks, which would help to increase the amount of energy absorbed during composite failure. This could provide a mechanism for increasing the value of $G_c$ for a composite. Hull [22, 23] has found a similar case when crushing fibre reinforced composite tubes; namely that brittle materials can have considerably higher absorption energies than ductile materials, because of extensive multiple microcracking in their failure zones.

Figs 8 and 9 show how the laminates have failed after three-point bending. With both types of laminate the pattern of visible cracks was similar, though these may not be responsible for the toughening mechanism, which probably requires multiple microcracking.

## 4.2. Interlaminar shear strength tests

From Table I it can be seen that the microstructure of the polypropylene phase affects the interlaminar shear strengths of the laminate composites. The measurements give an indication of the degree of adhesion between the sheets of glass and polypropylene. It appears in this case that the generation of transcrystalline layers has modestly increased the strength of the bond between the glass and the polypropylene. It has been reported that transcrystalline layers are prone to fracture close to their nucleating surfaces [2, 3, 9], but this did not adversely affect the results obtained here, in relation to the fine spherulitic material.

Our finding is different from that reported by previous workers. Some have found transcrystallinity to decrease adhesion [9, 10], while others have found that it makes little difference to adhesion [11, 12]. However, Schonhorn *et al.* [17] have predicted that if transcrystalline layers are relatively strong, this may be advantageous in achieving good adhesion; this appears to have been the case in our work.

Weak interfacial bonding can result in high values of $G_c$, by providing a crack deflection mechanism [24]

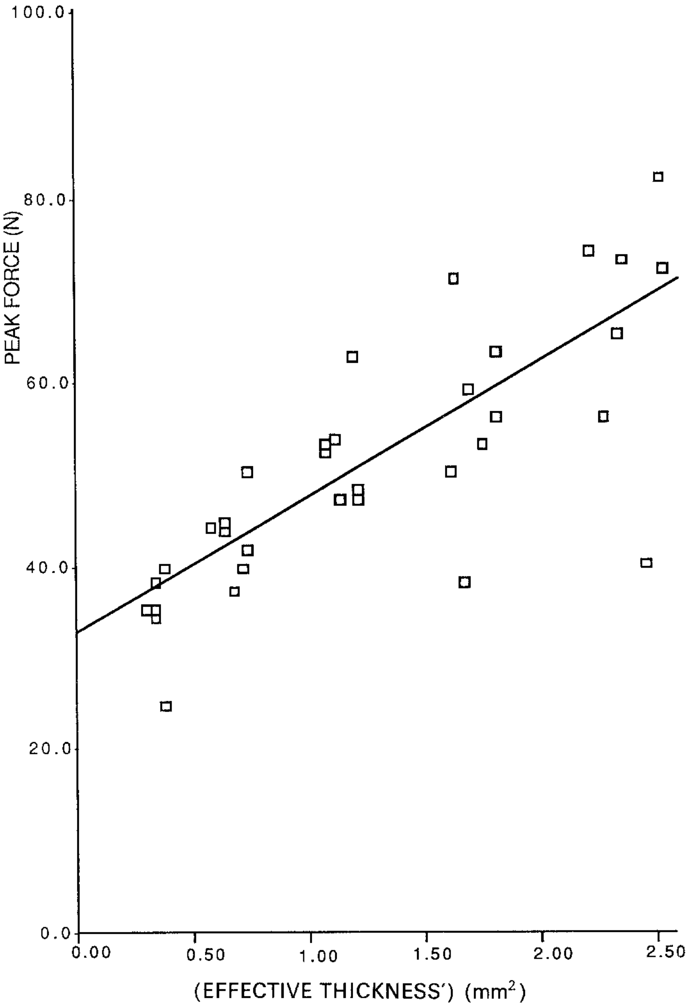

*Figure 6* Plot of failure peak force against the square of the effective thickness, for the determination of $G_c$ for a 6G7P laminate (with transcrystalline polypropylene).

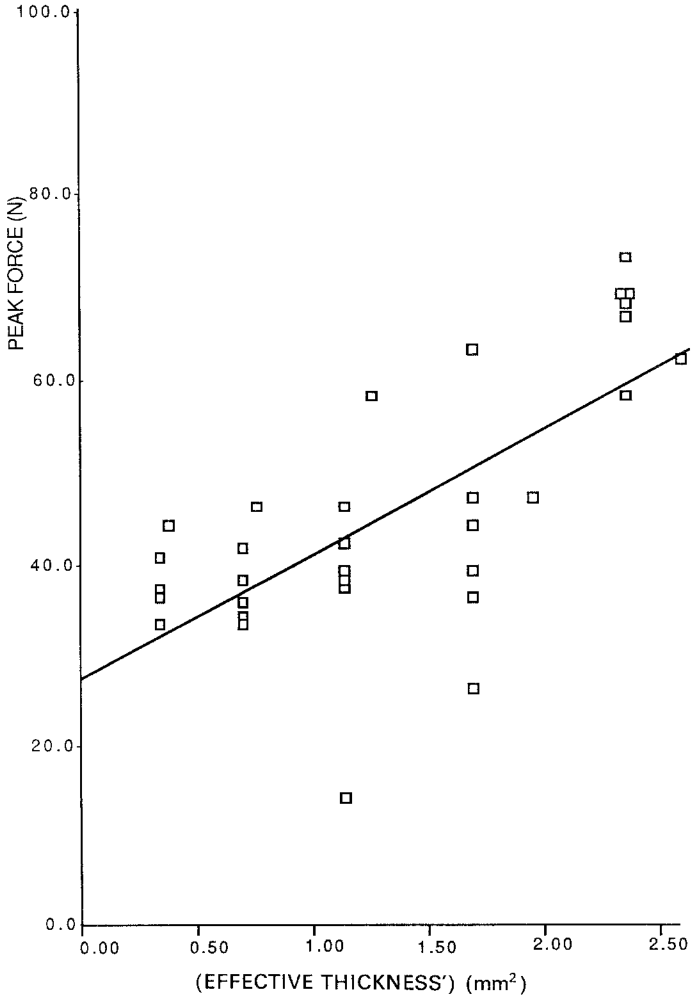

*Figure 7* Plot of failure peak force against the square of the effective thickness, for the determination of $G_c$ for a 6G7P laminate (with fine spherulitic polypropylene).

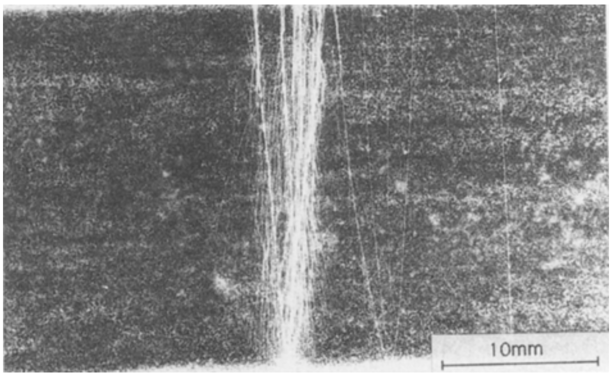

*Figure 8* Micrograph showing failure in a 6G7P laminate containing transcrystalline polypropylene, after a three-point-bending test.

between different layers. However, we have found that the generation of transcrystalline layers can actually lead to a higher degree of adhesion and higher values of $G_c$ compared with fine spherulitic morphologies. Because poor interfacial adhesion is more than likely to lead to a crack deflection process, it appears that an alternative toughening mechanism was occurring in those laminates tested in Section 4.1.

The differing effects of transcrystallinity on the interfacial bond strength for our laminates and for fibre-reinforced composites may well be related to the internal stress distribution resulting from specimen heat treatment. For example, for carbon-fibre-reinforced nylon 66 [9], thermal shrinkage stresses can result in fibre debonding, which can partially mask any effects due to interfacial morphology. Our laminates will be relatively unaffected in this way and thus the interlaminar shear strength measurements will provide a fairer assessment of the effects of interfacial microstructure compared with results derived from fibre-reinforced composites.

Examination of the fracture surfaces, using a scanning electron microscope (SEM), was undertaken and micrographs are shown in Figs 10 and 11. Fig. 10 shows a laminate containing transcrystallinity; the fracture has been a relatively brittle one. It is interesting to note that a crack appears roughly in the middle of the polypropylene layers, rather like interspherulitic fracture [25] and probably corresponds to where two transcrystalline layers meet. This indicates a zone of weakness, probably due to a lack of interconnecting material between two such layers. Previously it has been found that an area of failure occurred nearer a substrate (a fibre) or between transcrystalline layers or spherulites [2, 3, 9], rather than between transcrystalline layers themselves, as here. The fine spherulitic laminate (Fig. 11) by contrast shows a very ductile mode of failure.

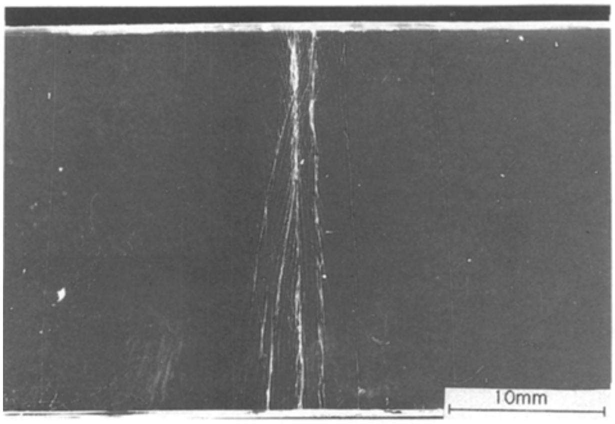

*Figure 9* Micrograph showing failure in a 6G7P laminate containing fine spherulitic polypropylene, after a three-point-bending test.

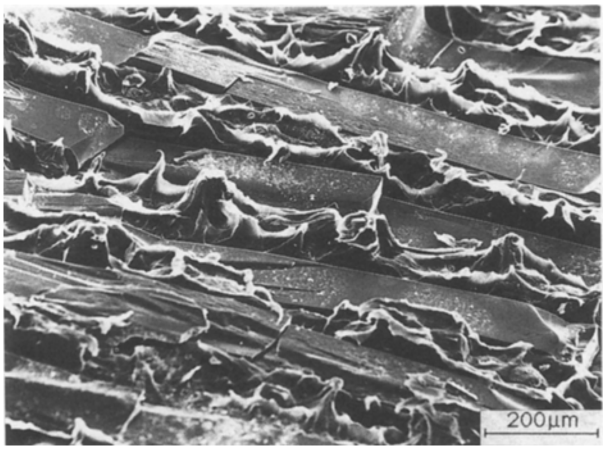

*Figure 10* SEM micrograph showing failure in a 14G15P laminate containing transcrystalline polypropylene, after a test for interlaminar shear strength.

The fact that the bonding was not particularly strong may have been a consequence of the specimen design. It has been found previously [26] that finely divided reinforcements, like fibres, require only relatively thin silane layers for effective adhesion; while massive structures, perhaps like the sheets used here, require relatively thick silane layers for effective adhesion. Possibly if thicker layers of silane had been put on the glass sheets, stronger adhesion may have been achieved.

## 4.3. Impact strengths

Values of $G_c$ (see Table II) may be obtained from the impact test data [19] using the equation

$$G_c = I_0/bdZ \tag{8}$$

where $Z$ is a geometric constant given by

$$Z = a/2d + S/18\pi a \tag{9}$$

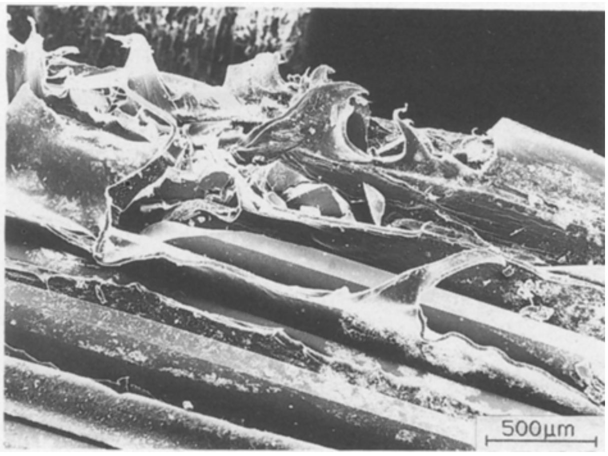

*Figure 11* SEM micrograph showing failure in a 14G15P laminate containing fine spherulitic polypropylene, after a test for interlaminar shear strength.

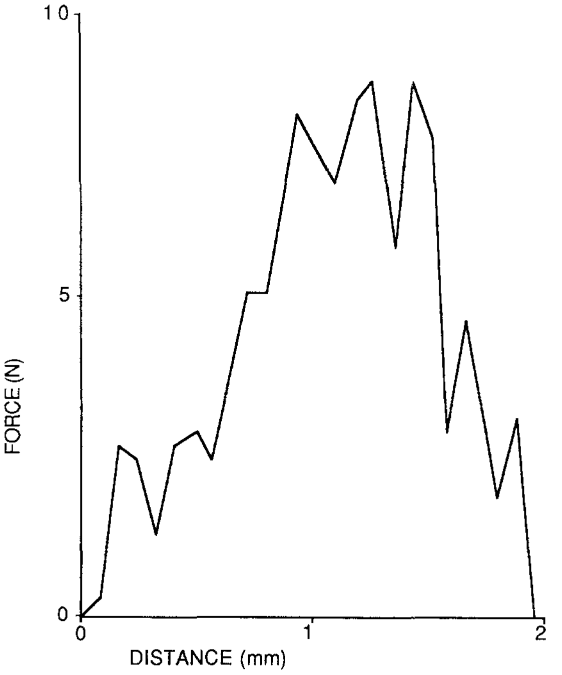

*Figure 12* Typical force/distance plot produced after a falling weight impact test on a fine spherulitic 6G7P laminate.

The data contained in Table I show that laminates containing transcrystalline polypropylene have slightly lower impact strengths than those containing fine spherulitic polypropylene. This was also reflected in the low energy-to-break values of transcrystalline polypropylene determined previously [16]. However, the $G_c$ values obtained from slow three-point bending would indicate that the generation of transcrystalline layers should increase the impact strength, but the conflicting results may be a reflection of the different testing rates used in the two measurement methods.

The force/distance plots obtained during impact testing, of which Fig. 12, was typical, show multiple failure peaks which could suggest cracking of individual layers in the composite. It is premature to attempt to relate the number of peaks on the force/distance plots to the number of layers of glass and/or polypropylene present, especially as it is widely known that some peaks can arise from multiple bouncing of the striker on the specimen. In fact the impact testing of single pieces of glass also tended to produce multiple (typically two to five) peaks on the force/distance plots obtained.

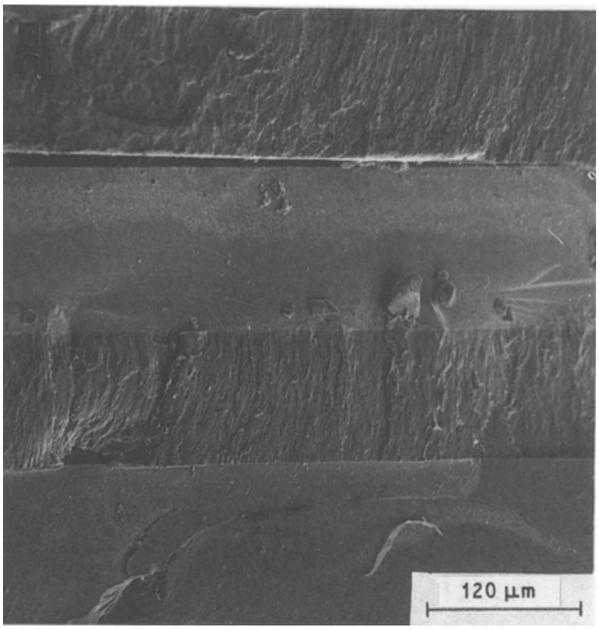

*Figure 13* SEM micrograph showing failure in a 6G7P laminate containing transcrystalline polypropylene, after a test for impact strength.

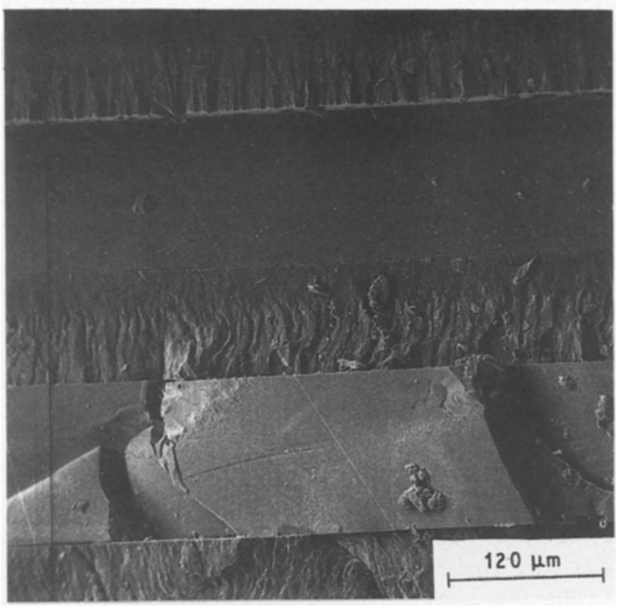

*Figure 14* SEM micrograph showing failure in a 6G7P laminate containing fine spherulitic polypropylene, after a test for impact strength.

Johnson *et al.* [27] have reported for glass fibre reinforced nylons that the distribution, number and size of the peaks on the force/distance plots can be varied by the use of different impact velocities and filtering devices. This adds further to the problems of interpreting these plots. However, it appears that multiple cracking could be an important feature of these sort of tests on some occasions, such as those with laminated composites.

The fracture surfaces of the two types of laminate were examined using the SEM, and are shown in Figs 13 and 14. The transcrystalline laminate, as one would expect, showed a more brittle fracture surface than the fine spherulitic one. Of particular interest were the distinctive fracture surfaces of the transcrystalline laminates, and the way that failure has occurred in the transcrystalline layers, leaving a very distinct texture.

## 5. Conclusions

The generation of transcrystalline layers in glass/polypropylene laminate composites causes increases in flexural strength and modulus, and can thus be said to have a reinforcing effect. This was consistent with the result obtained with polypropylene sheets [16].

One can observe the failure of individual layers in the laminates during three-point-bending tests, presumably because there are very distinct glass and polypropylene layers present; this was used to characterize the fracture properties of the laminates. It was found that the generation of transcrystalline layers caused an increase in the strain energy release rate, $G_c$. This result has not previously been reported in the literature; this appears to be the first time that the effects of transcrystallinity on the fracture performance of composites have been evaluated. This result is also consistent with the reports that transcrystalline zones may provide protective sheaths around fibres in composites.

A method of notching the laminates has recently been developed; this has yielded identical trends in $G_c$ to those reported here. These results will be published shortly.

The generation of transcrystalline layers appears slightly to harm impact strengths in the laminate composites. This may contradict the results obtained by three-point bending, but the speed of fracture used was very much faster.

The generation of transcrystalline layers has been found to increase the interlaminar shear strength of the laminate composites, so it may actually help to promote adhesion, though the effect is not large.

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