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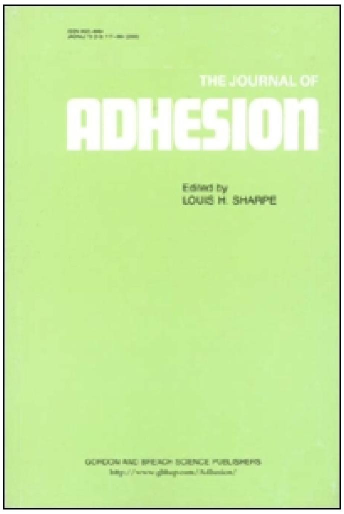

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The Adhesive Fracture Energy of Bonded Thermoplastic Fibre-Composites†

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The present paper first discusses the problems that occur when thermoplastic-based fibre-composite materials are bonded using structural engineering adhesives, such as epoxy and acrylic adhesives. A double-cantilever beam joint has been employed and it is shown that the value of the adhesive fracture energy, G_c, is very low when a simple abrasion/solvent wipe pretreatment is used for the thermoplastic fibre-composites. This arises from crack growth occurring along the adhesive/composite interface, which is relatively weak when such a pretreatment is employed. Secondly, it is demonstrated how very effective a corona surface pretreatment may be for these materials. Indeed, when such a pretreatment is used, interfacial crack growth is no longer observed but the crack now propagates either cohesively in the adhesive or through the composite substrate; both failure modes lead to relatively high values of G_c, with the former resulting in the highest values of G_c being recorded. Finally, from measuring the fracture properties of the composites and combining these data with a detailed analysis of the stresses in the DCB joint, calculated using a finite element analysis, the reasons for these different loci of failure may be readily understood and predicted.

INTRODUCTION

There is an increasing interest in the use of continuous carbon-fibre composites based upon thermoplastic matrices, rather than the well-established thermosetting matrix resins. This interest arises from several important advantages that thermoplastic polymeric matrices have to offer. The thermoplastic matrices commonly employed include a poly(ether-ether ketone) (PEEK), polyamide copolymer (PA), poly(ether imide) (PEI), polyimide (PI) and poly(phenylene sulphide) (PPS). The advantages that they offer include the ability readily to form the fibre-composite material into complex shapes and a greater resistance to impact damage due to the thermoplastic matrices usually being significantly tougher than the thermosetting matrices. However, some important problems have yet to be resolved before the properties of the thermoplastic-based

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composites may be fully exploited, and one such problem is how to adhesively-bond these composite materials successfully.

Previous work[1,2] has shown that, in the case of thermoplastic-based fibre-composite materials, the simple surface pretreatments commonly used for fibre-composites based upon thermosetting resins are insufficient to ensure adequate joint strengths when using structural engineering adhesives, which are typically based upon epoxy and acrylic resins. Thus, instead of simply using a light abrasion followed by a solvent-wipe for the surface pretreatment, as for thermosetting-resin composites, more complex surface pretreatments have been found to be necessary and have had to be developed. A previous publication by the authors first reported[2] the successful use of a corona treatment for a carbon-fibre/PEEK composite. The aims of the present paper are (i) to discuss the variation in the measured adhesive fracture energy, G_c, as a function of the corona treatment conditions for various thermoplastic composites bonded with different types of structural adhesives, and (ii) to show how the locus of failure of the bonded double-cantilever beam (DCB) joint used in the fracture mechanics tests may be predicted from a detailed stress analysis of the DCB joint and a knowledge of the transverse tensile properties of the fibre-composite substrate. It should be noted that the detailed surface, adhesion and stress analysis aspects of the present work will be presented in separate papers.[3,4]

EXPERIMENTAL

Fibre-composites

The fibre-composites studied consisted of nine different thermoplastic-based materials and one thermosetting-based composite, included for comparative purposes. These were:

(a) Unidirectional-carbon/PEEK ("APC-2" from ICI plc)—a continuous carbon-fibre composite containing a volume fraction of fibres of about 60% based upon a matrix of the thermoplastic polymer poly(ether-ether ketone). The composite substrate was prepared by laying unidirectional tape into a twelve-ply stack; the ply direction being $[0°]_{12}$. The moulding was carried out in a heated press at 380°C for 5 minutes and under a pressure of 1.4 MPa.

(b) Unidirectional-carbon/PA ("J2/Carbon" from Du Pont)—a continuous carbon-fibre composite containing a volume fraction of fibres of 55% based upon a matrix of a thermoplastic amorphous polyamide copolymer which is based on bis(para-amino cyclohexyl methane). The composite substrate was prepared by laying unidirectional tape into a twelve-ply stack; the ply direction being $[0°]_{12}$. The moulding was carried out in a heated press at 300°C for 25 minutes and under a pressure of 2 MPa.

(c) Woven-carbon/PA ("J2/Carbon" from Du Pont)—same as in (b) above except that the fibres were woven and the stack was made up of eleven plies.

(d) Unidirectional-"Kevlar"/PA ("J2/"Kevlar" " from Du Pont)—a continuous "Kevlar"-fibre composite containing a volume fraction of fibres of 60%

based upon a matrix of a thermoplastic amorphous polyamide copolymer which is based on bis(para-amino cyclohexyl methane). The composite substrate was prepared by laying unidirectional tape into a sixteen-ply stack; the ply direction being $[0°]_{16}$. The moulding was carried out in a heated press at 300°C for 25 minutes and a pressure of 2 MPa.

(e) Woven-"Kevlar"/PA ("J2/Kevlar"" from Du Pont)—same as in (d) above except that the fibres were woven and the stack was made up of nine plies.

(f) Woven-carbon/PEI ("CYPAC X7005" from American Cyanamid)—a woven carbon-fibre composite containing a volume fraction of fibres of about 62% based upon a matrix of a thermoplastic poly(ether imide). The composite substrate was prepared by laying the woven tape into an eight-ply stack. The moulding was carried out in a heated press at 315°C for 30 minutes and under a pressure of 0.7 MPa.

(g) Unidirectional-carbon/PPS ("AC40–60" from Phillips Petroleum)—a continuous carbon-fibre composite containing a volume fraction of fibres of about 53% based upon a matrix of the thermoplastic poly(phenylene sulphide). The composite substrate was prepared by laying unidirectional tape into a nine-ply stack; the ply direction being $[0°]_9$. The moulding was carried out in a heated press at 360°C for 5 minutes and under a pressure of 1.4 MPa.

(h) Unidirectional-carbon/PI ("JD861" from British Petroleum plc)—a continuous carbon-fibre composite containing a volume fraction of fibres of about 60% based upon the thermoplastic polyimide. The composite substrate was prepared by laying unidirectional tape into a nine-ply stack; the ply direction being $[0°]_9$. The moulding was carried out in a heated press at 300°C for 15 minutes under a pressure of 3.5 MPa.

(i) Woven-carbon/PI ("JD861" from British Petroleum plc)—same as in (h) above except that the fibres were woven and the stack was made of six plies.

(j) Unidirectional-carbon/epoxy ("913C XAS-5-34%" from Ciba Geigy Ltd)—a thermoset composite based upon a modified-epoxy supplied by Ciba-Geigy. It was a continuous carbon-fibre composite containing a nominal resin weight of about 34%. The composite substrate was prepared by laying unidirectional tap into a twelve-ply stack, the ply direction being $[0°]_{12}$. The moulding was carried out in a heated press at 150°C for 20 minutes under a pressure of 2.0 MPa. This thermoset composite was included for comparative purposes.

Adhesives

The structural adhesives employed were:

(a) A two-part, rubber-toughened, epoxy-paste adhesive ("EA 9309.3" from Hysol Dexter Corp.) which was cured at room temperature for 5 days under a pressure of 69 kPa.

(b) A rubber toughened epoxy-film adhesive ("FM73M" from American Cyanamid) which was cured at a temperature of 120°C for 1 hour under a pressure of 275 kPa.

(c) Two-part acrylic-paste adhesives ("F241", "F245" and "F246" from

Permabond Ltd.) which were cured at room temperature for 5 minutes under a pressure of 30 kPa.

(d) A two-part acrylic-paste adhesive ("V501" from Permabond Ltd.) which was cured at room temperature for 2 hours under a pressure of 30 kPa.

(e) Two part acrylic-paste adhesives ("M890" and "M896" from Bostik) which were cured at room temperature for 5 minutes under a pressure of 30 kPa.

To enable a comparison with the more common types of structural adhesives listed above the use of a hot-melt adhesive film, of the same chemical type as employed for the matrix of the thermoplastic composite, was also studied. The adhesive hot-melt films were:

(a) Poly(ether-ether ketone) (PEEK)—the PEEK hot-melt adhesive was applied to the corresponding composite substrate and the joints were prepared by subjecting the joint assembly to a temperature of 380°C for 5 minutes under a pressure of 0.5 MPa.

(b) Polyamide copolymer (PA)—the PA hot-melt adhesive was applied to the corresponding composite substrate and the joints were prepared by subjecting the joint assembly to a temperature of 300°C for 5 minutes under a pressure of 1 MPa.

(c) Poly(phenylene sulphide) (PPS)—the PPS hot-melt adhesive was applied to the corresponding substrate and the joints were prepared by subjecting the joint assembly to a temperature of 360°C for 5 minutes under a pressure of 1 MPa.

Substrate surface pretreatments

Several different surface pretreatments were conducted:

(a) Abrasion: here the composite sheets were lightly abraded using 180/220 mesh alumina, then wiped with methyl-ethyl ketone and allowed to dry.

(b) Moulding next to clean aluminium foil: to ensure no release agent was on the surface of the thermoplastic composite sheets were moulded against clean aluminium foil. The aluminium foil had previously been etched in a bath of chromic-acid solution to ensure that there were no release-agents on its surface. After the moulding operation the aluminium foil was dissolved away by using a bath of two molar sodium hydroxide solution.

(c) Acid etch: thermoplastic composite specimens were etched in an acidic solution[5] based upon 1% solution of potassium permanganate in a 5:2:2 solution of sulphuric acid, orthophosphoric acid and distilled water.

(d) Corona discharge: the major components of the corona discharge equipment were the generator producing high frequency (15–20 kHz) power (0.1–0.9 kW), the high-power transformer giving the high voltage (15–20 kV) and the high-power cables carrying the high-voltage to the electrodes and the treater-station. The equipment was designed to include two special features. Firstly, conventional corona can readily treat non-conducting materials but the transformer of the present equipment was redesigned to give good impedance and capacitance matching between the electrode and the composite. For conducting materials a modified electrode was designed with a silicone rubber covering the surface whilst for non-conducting materials a conventional knife-edge electrode was used. The second feature was that the power output from the electrode was current controlled and not voltage controlled. Hence the power output from the electrode was directly obtained from the power gauge of the generator. After a light abrasion and a solvent wipe treatment, which is described above but is not a critical requirement, the thermoplastic-composite substrates were placed on an automatically-controlled table which travelled horizontally backwards and forwards under the discharge electrode. The velocity of the table could be selected to be between 14.5 to 62 mm/s and the velocity was controlled accurately by a stepper motor and a pulse generator (0.1–4.8 kHz). The energy output per unit area from the electrode onto the composites may be determined from:

$$E = PN/LV \tag{1}$$

where E is the energy output per unit area, P is the power of the high-frequency generator, N is the number of cycles of the table, L is the length of the treater and V is the velocity of the table.

Joint preparation

After preparing the composite sheets they were cut to equal strips and an appropriate pretreatment was applied. Then an adhesive was chosen and applied on one side of each of two composite substrates to make a double-cantilever-beam (DCB) joint specimen. After applying the adhesive a release-coated aluminium foil was inserted in between the adhesive-coated composite substrates to act as a starter crack. The specimen details were as follows:

Length of composite substrate	L	140 mm
Width of composite substrate	B	20 mm
Thickness of each composite substrate	t	1.5 mm
Length of crack starter	a_0	20 mm
Typical thickness of the cured adhesive	t_a	0.4 mm

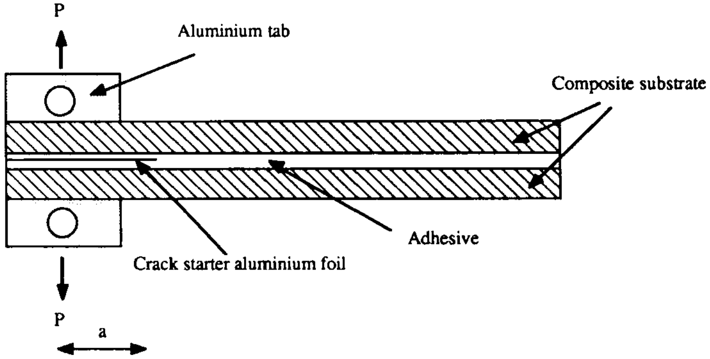

FIGURE 1 The double-cantilever beam (DCB) joint test.

Note that for each specimen bonded with the hot-cure epoxy-film (FM73M) adhesive two layers of this film-adhesive were used.

After the adhesives had been cured, as described above, two aluminium tabs, 20 × 20 × 10 mm in size, were bonded onto the DCB joints. The adhesive used to bond the aluminium tabs was a room curing epoxy-paste adhesive ("E38" from Permabond). Afterwards the edges of the DCB specimens were painted white using a typewriter correction fluid and were marked off at 0.5 cm intervals along the complete length of the beam; see Figure 1 for details.

DCB joint testing and analysis

The DCB joint specimens were mounted in a screw-driven tensile-testing machine and loaded under displacement control at a crosshead speed of 2 mm/min. The tensile-testing machine was connected to a computer which recorded the load *versus* displacement trace. The crack was monitored using a travelling microscope mounted on a stand and, as the crack propagated and crossed the markers, a "bleeper" on the tensile-testing machine were used to indicate the corresponding crack lengths on the load-displacement trace. The equation used to calculate the adhesive fracture energy, G_c, was:

$$G_c = (P_c^2/2B)(\partial C/\partial a) \tag{2}$$

and $C = \Delta/P$ where

P_c = load for crack growth
P = load
B = width
C = compliance of the specimen
Δ = displacement
a = crack length

All the variables mentioned above were directly measured using the experimental procedure described above. To evaluate the partial derivative of the compliance with a crack length a special computer program was developed to curve-fit the experimental data of the compliance *versus* the crack length. An important feature of this software was that it curve-fitted the experimental data such that as $a \rightarrow 0$ then $\partial C/\partial a \rightarrow 0$.

Interlaminar fracture energy, G_c (il), measurements

The interlaminar fracture energy, G_c(il), of the composites was also measured. For these measurements specimens were prepared which were 140 × 20 × 1.5 mm in size and which contained a short length of release-coated aluminium foil between the centre plies. Aluminium end-tabs were bonded onto the interlaminar specimen so that it could be loaded using the tensile testing machine, which was again driven at a cross head speed of 2 mm/min. The steady-state propagation value of the interlaminar fracture energy, G_c(il), was calculated using Eq. (2).

TABLE I
The out-of-plane transverse tensile stresses to failure, σ_{yyc}, for different composites

Composite	Transverse tensile stress σ_{yyc} (MPa)
Unidirectional-carbon/PEEK	84.3 ± 4.1
Unidirectional-carbon/PA	83.7 ± 4.2
Woven-carbon/PA	83.7 ± 4.2
Unidirectional "Kevlar"/PA	21.5 ± 2.9
Woven "Kevlar"/PA	21.5 ± 2.9
Unidirectional-carbon/PPS	29.4 ± 3.6
Unidirectional-carbon/PI	56.1 ± 3.4
Woven-carbon/PI	56.1 ± 3.4
Unidirectional-carbon/epoxy	58.9 ± 3.3

Note: Crosshead-displacement rate: 1 mm/min.

Transverse tensile properties of the composite materials

In order to predict quantitatively the locus of failure which was observed for the different DCB specimens it was found necessary to assess the transverse tensile strength, σ_{yyc}, of the various composite materials. The recommendations of ASTM Standard D-3039 were followed and specimens of $125 \times 25 \times 1.5$ mm were tested with the fibre direction being 90° with respect to the loading direction. Five such specimens were tested for each type of composite. It should be noted that in these studies two assumptions have been made. Firstly, that the transverse tensile strength of the unidirectional composites is the same in the planes perpendicular to the reinforced plane and, secondly, that the transverse tensile strength is the same for woven and unidirectional composites provided the fibre type and the matrix resin are the same.

The results from such tests are shown in Table I. As will be discussed in detail later, the transverse tensile stresses, σ_{yyc}, required for failure of the composites were there was good adhesion between the fibres and the matrix polymer were relatively high, whereas for the composites where the adhesion between the fibres and the matrix was poor then the values of σ_{yyc} were relatively low.

Fractography

The fracture surfaces of the different test specimens were examined in the scanning electron microscope, after the surface had been vacuum-coated with a thin layer of gold to avoid charging effects.

RESULTS AND DISCUSSIONS

Values of G_c for the carbon-fibre/eopxy bonded joints

Before considering the results for the thermoplastic composites it is of interest to consider the values of the adhesive fracture energy, G_c, which were obtained for

TABLE II
Adhesive fracture energy, G_c, and locus of failure data for unidirectional carbon-fibre/epoxy composite

Substrate pretreatment	Adhesive	Adhesive type	G_c (kJ/m^2)	Locus of failure
Abrasion/solvent wipe	EA93093	2-part epoxy	0.25	Interlaminar
Abrasion/solvent wipe	FM73M	1-part epoxy	1.90	Cohesive
Abrasion/solvent wipe	F241	2-part acrylic	1.30	Cohesive
Abrasion/solvent wipe	F245	2-part acrylic	1.50	Cohesive
Abrasion/solvent wipe	F246	2-part acrylic	1.45	Cohesive
Abrasion/solvent wipe	V501	2-part acrylic	0.90	Cohesive
Abrasion/solvent wipe	M890	2-part acrylic	0.70	Cohesive
Abrasion/solvent wipe	M896	2-part acrylic	0.75	Cohesive

Note: (i) "Cohesive" means joint failure occurred by cohesive fracture in the adhesive layer. (ii) "Interlaminar" means joint failure occurred in an interlaminar fracture in the composite substrate.

the thermosetting composite, which was a unidirectional carbon-fibre/epoxy composite. Such results obviously act as a starting point when considering later the adhesive bonding of the thermoplastic-based composites. Following a simple light abrasion and solvent wipe pretreatment, the values of G_c and locus of failure which were recorded are shown in Table II.

As may be seen from the data shown in Table II, the simple abrasion/solvent wipe pretreatment employed for the epoxy-based composite has been sufficient to ensure good wetting and adhesion of the adhesive to the composite substrates to that extent that the joints typically fail by the starter crack, which was inserted in the adhesive layer, continuing to propagate cohesively through the adhesive layer or, in one instance, by a new crack initiating and growing in the composite substrate to give an interlaminar locus of failure. In the latter instance the value of the measured adhesive fracture energy, G_c, is equal in value to that of the interlaminar fracture energy, G_c(il), of the epoxy fibre-composite, as discussed below and shown in Table VII.

Thus, the observation that, for a freshly-prepared thermosetting-based composite, a simple abrasion/solvent wipe pretreatment is sufficient to avoid any indications of failure at the adhesive/composite interface is in accord with previously published results.[6]

Values of G_c for the carbon-fibre/PEEK bonded joints

The preliminary general studies were largely conducted using the unidirectional carbon-fibre/poly(ether-ether ketone) laminate. The initial results which were obtained are shown in Table III below.

Several conclusions may be drawn from the data shown in Table III. Firstly, a simple abrasion/solvent wipe pretreatment is clearly inadequate for the carbon-fibre/PEEK thermoplastic composite when the epoxy or acrylic adhesives are employed. Secondly, neither the aluminium foil/NaOH nor the acid-etch

TABLE III
Adhesive fracture energy, G_c, and locus of failure data for unidirectional carbon-fibre/PEEK composite subjected to various pretreatments and bonded with different adhesives

Substrate pretreatment	Adhesive	Adhesive type	G_c (kJ/m^2)	Locus of failure
Abrasion/solvent wipe	EA9309.3	2-part epoxy	0.03	Interfacial
Abrasion/solvent wipe	FM73M	1-part epoxy	0.02	Interfacial
Abrasion/solvent wipe	F241	2-part acrylic	0.01	Interfacial
Abrasion/solvent wipe	F245	2-part acrylic	0.02	Interfacial
Abrasion/solvent wipe	F246	2-part acrylic	0.03	Interfacial
Abrasion/solvent wipe	V501	2-part acrylic	0.02	Interfacial
Abrasion/solvent wipe	M890	2-part acrylic	0.03	Interfacial
Abrasion/solvent wipe	M891	2-part acrylic	0.03	Interfacial
Abrasion/solvent wipe	PEEK	Hot-melt	2.25	See note (iv)
Al foil/NaOH	EA9309.3	2-part epoxy	0.12	Interfacial
Al foil/NaOH	FM73M	1-part epoxy	0.40	Interfacial
Acid etch, 5 min	EA9309.3	2-part epoxy	0.30	Interfacial
Acid etch, 10 min	EA9309.3	2-part epoxy	0.90	Interfacial
Acid etch, 5 min	FM73M	1-part epoxy	1.30	Mainly interfacial
Acid etch, 10 min	FM73M	1-part epoxy	1.65	Mainly cohesive

Notes: (i) "Cohesive" means cohesive in the adhesive layer. (ii) "Interfacial" means joint failure occurred along the adhesive/composite interface. (iii) "Interlaminar" means joint failure occurred between the laminae in the composite substrate. (iv) Either cohesive or interlaminar; but due to the thinness of the layer of PEEK hot-melt film used the exact locus of failure could not be ascertained.

treatments are very effective for the cold-cured epoxy paste adhesive ("EA9309.3"), although both treatments are somewhat more effective in the case of the hot-cured epoxy film adhesive ("FM73M"); this possibly arises from the improved wetting likely to occur when the hot-cured adhesive is employed. These observations suggest that the presence on the carbon-fibre/PEEK composite of a weak boundary layer, for example due to release agents, is unlikely to be the essence of the problem. Thirdly, this conclusion is supported by the ability of the PEEK to act as a most effective hot-melt adhesive for the carbon-fibre/PEEK composite. Thus, a possible cause for the interfacial failure recorded for the carbon-fibre/PEEK composite joints, which results in low G_c values, is the poor wettability and intrinsic adhesion of the thermoplastic composite surface relative to the epoxy or acrylic adhesives. Surface analysis studies[2,3] using X-ray photoelectron spectroscopy have confirmed the above conclusions and these results, together with the detailed mechanisms of adhesion, will be reported in a separate publication.[4]

The above conclusion suggests that chemical modification of the surface of the thermoplastic composite by introducing specific groups which will increase the degree of interfacial contact and intrinsic adhesion may be beneficial. Therefore, the corona-discharge pretreatment was employed, since this is known frequently to increase the wettability and adhesion of "difficult-to-bond" substrates. The results are shown in Table IV for the epoxy adhesives and in Table V for the acrylic adhesives; note that all the results for the acrylic adhesives were similar so

TABLE IV
Effects of a corona-discharge pretreatment on the values of G_c and the observed locus of failure (L of F) for the unidirectional carbon-fibre/PEEK composite bonded with the epoxy adhesives

Treatment level	EA9309.3 adhesive		FM73M adhesive	
($\mathrm{J/mm^2}$)	G_c ($\mathrm{kJ/m^2}$)	L of F	G_c ($\mathrm{kJ/m^2}$)	L of F
0	0.03	Interfacial	0.02	Interfacial
0.125	0.03	Interfacial	0.02	Interfacial
0.25	0.02	Interfacial	0.12	Interfacial
0.5	0.04	Interfacial	0.17	Interfacial
0.75	0.09	Interfacial	0.18	Interfacial
1.0	0.11	Interfacial	0.54	Interfacial
2.5	0.36	Interfacial	1.22	Interfacial + Cohesive
5.0	0.96	Interfacial	1.44	Interfacial + Cohesive
10.0	1.74	Interfacial + Cohesive	1.84	Cohesive
15.0	3.12	Interfacial + Cohesive	1.82	Cohesive
20.0	3.72	Cohesive	1.84	Cohesive
25.0	3.88	Cohesive	1.82	Cohesive
30.0	3.90	Cohesive	1.86	Cohesive

only a few typical results have been included in Table V. As may be seen, the corona treatment may indeed lead to an increase in the value of G_c for the thermoplastic PEEK fibre-composite bonded with the epoxy- or acrylic-based adhesives. Further, as expected, the higher values of G_c are associated with the joints failing now by cohesive fracture through the adhesive layer, rather than along the composite/adhesive interface.

The data given in Table IV for the epoxy adhesives are also plotted in Figure 2 in the form of the measured adhesive fracture energy, G_c, *versus* the level of corona pretreatment employed for the unidirectional carbon-fibre/PEEK composite (again expressed as an output energy level, E, per unit area of composite surface). There are several interesting points to note. Firstly, for both adhesives the value of G_c increases steadily as the treatment level is increased until a plateau is reached. Once the maximum, plateau value of G_c is reached then it appears that an increased level of corona treatment offers no further advantages to the performance of the joint. The plateau values of the adhesive fracture energy, G_c, are about 2 and 4 $\mathrm{kJ/m^2}$ for the joints bonded using the hot-cured

TABLE V
Effects of a corona-discharge pretreatment on the values of G_c and the observed locus of failure (L of F) for the unidirectional carbon-fibre/PEEK composites bonded with the acrylic-based adhesives

Treatment level	F241 adhesive		V501 adhesive		M890 adhesive	
($\mathrm{J/mm^2}$)	G_c ($\mathrm{kJ/m^2}$)	L of F	G_c ($\mathrm{kJ/m^2}$)	L of F	G_c ($\mathrm{kJ/m^2}$)	L of F
0	0.02	Interfacial	0.05	Interfacial	0.05	Interfacial
20.0	1.25	Cohesive	0.97	Cohesive	0.67	Cohesive

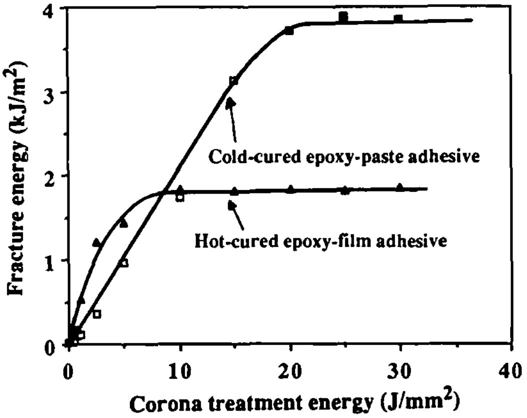

FIGURE 2 The adhesive fracture energy, G_c, *versus* the corona-treatment energy level for the unidirectional carbon-fibre/poly(ether ether ketone) (PEEK) composite bonded with the cold-cured epoxy-paste (EA9309.3) and the hot-cured epoxy-film (FM73M) adhesives. (Open points: interfacial failure; closed points: cohesive failure in adhesive).

epoxy-film adhesive (FM73M) and the cold-cured epoxy-paste (EA9309.3) adhesive, respectively. Secondly, as might be expected, it is only when the treatment level is sufficient to give an intrinsically strong interface that cohesive failure through the adhesive is observed, as opposed to the interfacial locus of failure which is observed at low pretreatment levels. Thirdly, again as expected, the treatment level needed to result in the joint failing by cohesive fracture through the adhesive is very similar to that level needed to give the maximum, plateau, value of G_c. Fourthly, the level of corona pretreatment needed for the composite joint to attain the maximum value of G_c is dependent upon the adhesive being employed: a tougher adhesive, having the potential to give a greater value of the adhesive fracture energy, G_c, requires a higher level of corona treatment in order to achieve the maximum crack resistance for the joint than a less-tough adhesive.

Values of G_c for the other thermoplastic fibre-composites

Firstly, for all the other thermoplastic composites examined in the present studies it has also been found that they may be bonded successfully using a film of the matrix polymer as a hot-melt adhesive. The results are shown in Table VI. It is of interest to note that the values of the adhesive fracture energy, G_c, are approximately the same as for the interlaminar fracture energy, G_c(il), see Table VII.

Secondly, however, if any of the thermoplastic composites are subjected simply to an abrasion/solvent-wipe pretreatment and then bonded using the epoxy structural adhesives the value of G_c obtained is extremely low with the locus of failure being along the adhesive/composite interface, just as observed for the

TABLE VI
Adhesive fracture energy, G_c, for the thermoplastic composites bonded using a film of the matrix polymer as a hot-melt adhesive

Composite	Adhesive	G_c (kJ/m^2)	Locus of failure
Unidirectional carbon-fibre/PA	PA	1.05	Cohesive or Interlaminar
Woven carbon-fibre/PA	PA	1.15	Cohesive or Interlaminar
Unidirection Kevlar-fibre/PA	PA	0.60	Cohesive or Interlaminar
Woven Kevlar-fibre/PA	PA	0.60	Cohesive or Interlaminar
Unidirectional carbon-fibre/PPS	PPS	1.45	Cohesive or Interlaminar

Note: (i) The locus of joint failure was either cohesive or interlaminar; but due to the thinness of the layer of hot-melt films used the exact locus of failure could not be ascertained.

PEEK fibre-composites and described above. Hence, again there appears to be a problem in attaining sufficient wetting and intrinsic adhesion at the epoxy-adhesive/composite interface.

Thirdly, therefore, the various thermoplastic fibre-composites were subjected to the corona-discharge pretreatment and the results are shown in Figures 3 to 10. The data for the different fibre-composite/adhesive combinations are discussed below.

(i) The results shown in Figures 3 and 4 for the unidirectional and woven carbon-fibre/PA thermoplastic composites are very similar in form to those shown in Figure 2 for the PEEK composite. Namely, the value of G_c rises to a maximum, plateau value as the level of corona pretreatment used prior to bonding is increased, and this effect is accompanied by a change in the locus of joint failure from interfacial to cohesive in the adhesive. However, note that the

TABLE VII
Joint and composite failure properties

	Data for DCB Joints—adhesive employed:				
	Cold-cured epoxy (EA9309.3)		Hot-cured epoxy (FM73M)		Composite
Composite	G_c (plateau)	L of F	G_c (plateau)	L of F	G_c(il)
U/d carbon/epoxy	0.25	Il	1.90	Coh	0.25
U/d carbon/PEEK	3.90	Coh	1.86	Coh	2.30
U/d carbon/PA	3.78	Coh	1.79	Coh	1.10
Woven carbon/PA	3.84	Coh	1.84	Coh	1.15
U/d Kevlar/PA	0.64	Il	0.66	Il	0.65
Woven Kevlar/PA	0.71	Il	0.70	Il	0.68
U/d carbon/PI	1.51	Il	1.50	Il	1.46
Woven carbon/PI	1.69	Il	1.86	Coh	1.68
Woven carbon/PEI	1.67	Il	1.84	Coh	1.60
U/d carbon/PPS	1.47	Il	1.47	Il	1.45

Notes: (i) "G_c (plateau)" is the plateau value of the adhesive fracture energy which, for the thermoplastic composites, occurs after a given corona pretreatment level; obviously this is the uniform constant value for the carbon-fibre/epoxy composite since no corona treatment is needed for this composite to ensure high G_c values and cohesive fracture. (ii) "G_c(il)" is the interlaminar fracture energy for the composite. (iii) All G_c values are in kJ/m^2. (iv) "U/d": unidirectional. (v) "Coh": cohesive in adhesive layer failure; "Il": interlaminar fracture in composite.

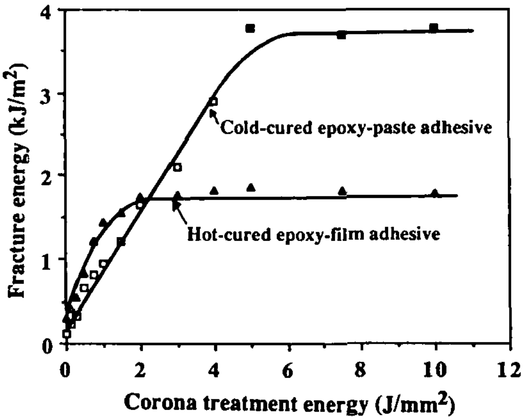

FIGURE 3 The adhesive fracture energy, G_c, *versus* the corona-treatment energy level for the unidirectional carbon-fibre/polyamide copolymer (PA) composite bonded with the cold-cured epoxy-paste (EA9309.3) and the hot-cured epoxy-film (FM73M) adhesive. (Open points: interfacial failure; closed points: cohesive failure in adhesive).

level of corona treatment needed to attain the plateau value of G_c is significantly less than that required for the PEEK composite, reflecting the higher initial polarity of the amorphous polyamide copolymer, which is based on bis(para-amino cyclohexyl methane), compared to the poly(ether-ether ketone) matrix.

(ii) Figures 5 and 6 show the values of G_c *versus* the level of corona treatment employed for the unidirectional "Kevlar"-fibre/PA composites and the woven "Kevlar"-fibre/PA composites, respectively. Note in both cases the corona treatment is effective in increasing the adhesive fracture energy but with both composites, and using either the cold-cured epoxy-paste (EA9309.3) or hot-cured

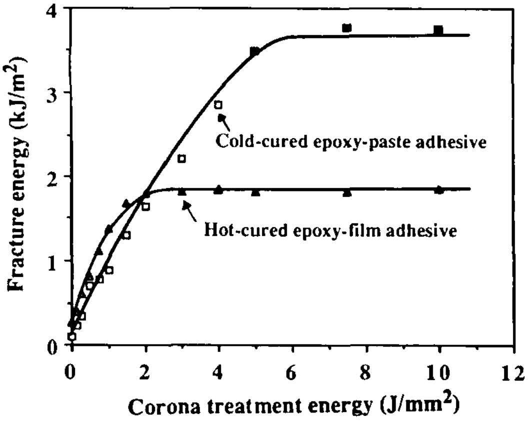

FIGURE 4 The adhesive fracture energy, G_c, *versus* the corona-treatment energy level for the woven carbon-fibre/polyamide copolymer (PA) composite bonded with the cold-cured epoxy-paste (EA9309.3) and the hot-cured epoxy-film (FM73M) adhesives. (Open points: interfacial failure; closed points: cohesive failure in adhesive).

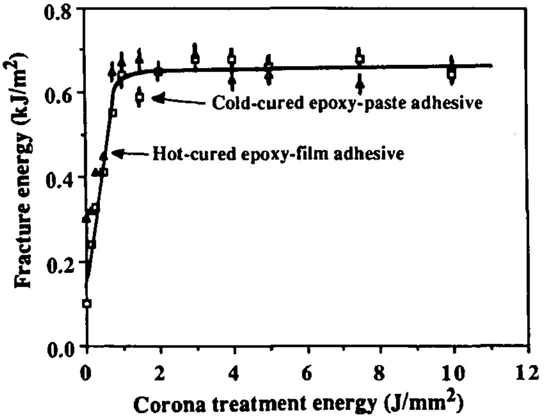

FIGURE 5 The adhesive fracture energy, G_c, *versus* the corona-treatment energy level for the unidirectional "Kevlar"-fibre/polyamide copolymer (PA) composite bonded with the cold-cured epoxy-paste (EA9309.3) and the hot-cured epoxy-film (FM73M) adhesives. (Squares EA9309.3; triangles: FM73M; open points: interfacial failure; open points with vertical bar: interlaminar failure of the composite substrate).

epoxy-film (FM73M), the plateau value of G_c is relatively low; well below the values observed in Figures 3 and 4, for example. Thus, the plateau values of G_c obtained for the "Kevlar"-fibre/PA composites are significantly lower than those obtained for the carbon-fibre/PA composites. The reason for this may be readily understood by observing that under these circumstances the locus of joint failure for the "Kevlar"-fibre/PA composites bonded using the cold-cure epoxy-paste (EA9309.3) and hot-cured epoxy-film (FM73M) adhesives is by an interlaminar fracture through the composite, *i.e.* the composite substrate delaminates by a new

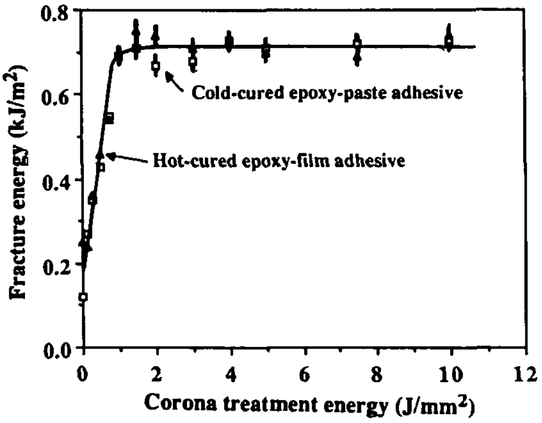

FIGURE 6 The adhesive fracture energy, G_c, *versus* the corona-treatment energy level for the woven "Kevlar"-fibre/polyamide copolymer (PA) composite bonded with the cold-cured epoxy-paste (EA9309.3) and the hot-cured epoxy-film (FM73M) adhesives. (Squares EA9309.3; triangles: FM73M; open points: interfacial failure; open points with vertical bar: interlaminar failure of the composite substrate).

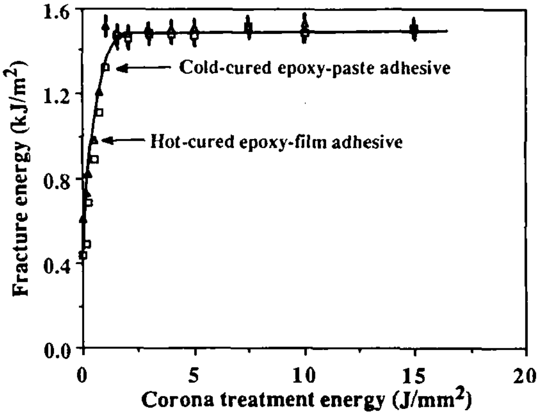

FIGURE 7 The adhesive fracture energy, G_c, *versus* the corona-treatment energy level for the unidirectional carbon-fibre/thermoplastic polyimide (PI) composite bonded with the cold-cured epoxy-paste (EA9309.3) and the hot-cured epoxy-film (FM73M) adhesives. (Squares EA9309.3; triangles: FM73M; open points: interfacial failure; open points with vertical bar: interlaminar failure of the composite substrate).

crack initiating and propagating between the laminae forming the composite. The premature interlaminar failure of the composite substrate prevents the far-higher values of G_c associated with cohesive fracture through the tough adhesives from being obtained. Thus, whilst the corona pretreatment has been effective in preventing interfacial failure along the adhesive/composite interface, the "weak link" in the joint is now the premature delamination of the substrate itself.

(iii) The data for the unidirectional carbon-fibre/thermoplastic polyimide, woven carbon-fibre/thermoplastic polyimide, woven carbon-fibre/poly(ether

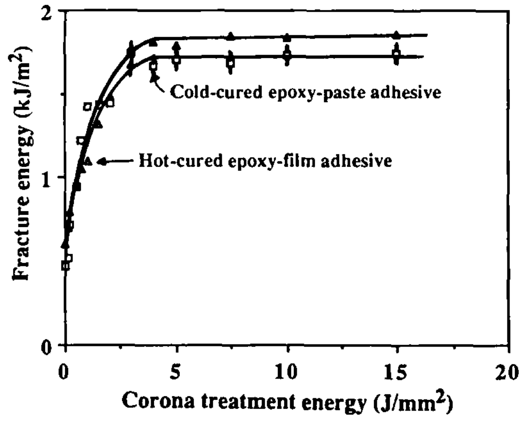

FIGURE 8 The adhesive fracture energy, G_c, *versus* the corona-treatment energy level for the woven carbon-fibre/thermoplastic polyimide (PI) composite bonded with the cold-cured epoxy-paste (EA9309.3) and the hot-cured epoxy-film (FM73M) adhesives. (Squares EA9309.3; triangles: FM73M; open points: interfacial failure; closed points: cohesive failure in adhesive; open points with vertical bar: interlaminar failure of the composite substrate).

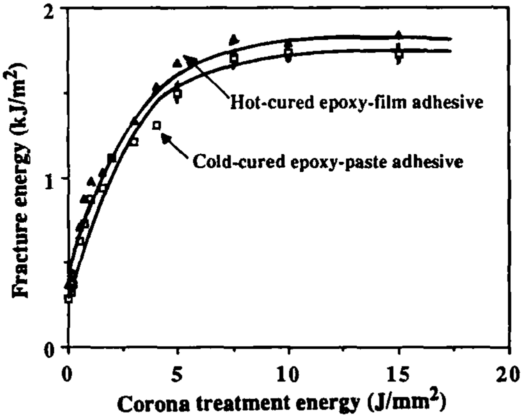

FIGURE 9 The adhesive fracture energy, G_c, *versus* the corona-treatment energy level for the woven carbon-fibre/poly(ether imide) (PEI) composite bonded with the cold-cured epoxy-paste (EA9309.3) and the hot-cured epoxy-film (FM73M) adhesives. (Squares EA9309.3; triangles: FM73M; open points: interfacial failure; closed points: cohesive failure in adhesive; open points with vertical bar: interlaminar failure of the composite substrate).

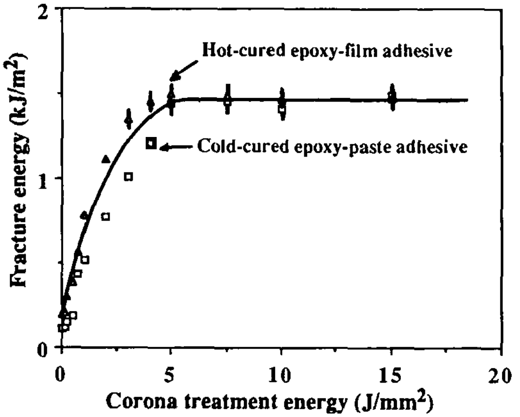

FIGURE 10 The adhesive fracture energy, G_c, *versus* the corona-treatment energy level for the unidirectional carbon-fibre/thermoplastic poly(phenylene sulphide) (PPS) composite bonded with the cold-cured epoxy-paste (EA9309.3) and the hot-cured epoxy-film (FM73M) adhesives. (Squares EA9309.3; triangles: FM73M; open points: interfacial failure; open points with vertical bar: interlaminar failure of the composite substrate).

imide) and unidirectional carbon-fibre/poly(phenylene sulphide) composites are given in Figures 7, 8, 9 and 10, respectively, and a similar picture to that described in the preceding paragraphs emerges.

Locus of failure studies

Introduction The above studies have clearly shown that for the thermosetting epoxy fibre-composite a simple abrasion/solvent wipe pretreatment of the composite is all that is needed to ensure a good interfacial strength with the bonded joints failing by cohesive fracture through the adhesive layer at a high value of the adhesive fracture energy, G_c. In the case of the thermoplastic composites a corona pretreatment is necessary to prevent the structural adhesive/composite interface failing at a very low value of G_c. Also, after the thermoplastic composite has been subjected to a certain level of corona treatment the value of G_c reaches a maximum, plateau value. However, whilst in some cases this does correspond to a cohesive fracture through the adhesive layer at a high value of G_c, in other cases the "weak link" now appears to be the composite substrate itself. In these instances the bonded DCB joint fails by a new crack initiating between the laminae in the composite, very close to the adhesive/composite interface, and just above and/or below the starter crack placed in the adhesive layer. This interlaminar crack propagates through the composite substrate resulting in joint fracture. The important practical consideration is that such an interlaminar crack propagates at an applied load, and hence an adhesive fracture energy, G_c, below that associated with cohesive failure through the adhesive layer.

The above points may be clearly seen from the data shown in Table VII. Firstly, note that when the bonded joint fails by interlaminar fracture of the composite substrate the associated value of the maximum adhesive fracture energy, G_c (plateau), attained is significantly lower than when the locus of joint failure is *via* cohesive fracture through the adhesive layer. Indeed, when the joints fail by interlaminar fracture of the composite the measured value of G_c is very similar to that of G_c(il), as would be expected. Hence, using the present tough adhesives, this leads to the measured value of G_c plateau being, in some instances, only about 15 to 20% of what would be attainable if delamination of the thermoplastic composite did not intervene as the operative failure mechanism. Secondly, as might be expected, there is a greater tendency for the joints employing the cold-cured epoxy-paste adhesive (EA9309.3) to exhibit an interlaminar failure of the composite substrate. This is because the cold-cured epoxy-paste adhesive (EA9309.3) is the tougher of the two epoxy adhesives used and, hence, higher loads need to be applied to the joint to cause crack growth through the adhesive layer. These higher loads will give rise to higher stresses in the composite substrate. Therefore, the composite may be able to withstand delamination when bonded with the less tough hot-cured epoxy-film (FM73M) adhesive but will delaminate when subjected to the higher loads and stresses which can be imposed when the tougher cold-cured adhesive is employed. Thirdly, there appears to be little correlation between the interlaminar fracture energy, G_c(il), of the composite and the propensity of the adhesive joint to fail by interlaminar failure of the composite substrate. For example, the epoxy-based composite has by far the lowest G_c(il) value, but when bonded using the hot-cured (FM73M) adhesive the joints show no signs of any interlaminar failure. Also, the unidirectional or the woven carbon-fibre/PA thermoplastic composites possess only a moderate value of G_c(il), but again DCB joints using these composites show no signs of any interlaminar fracture. Thus, it appears that some other property of the composite must control its tendency to delaminate when bonded and tested as a DCB joint.

Out-of-plane transverse tensile fracture stress, σ_{yyc}, of the composite materials The obvious property to investigate is the out-of-plane transverse tensile fracture stress, σ_{yyc}, of the composite materials, since a new crack must form in the composite substrates by a transverse tensile fracture between the laminae. This property was measured as described earlier and the results are shown in Table I. The mean values of σ_{yyc} for the various composites are shown in Table VIII and, as may be seen, there is a good qualitative agreement between the values of σ_{yyc} and the propensity of the bonded DCB joint to exhibit an interlaminar failure mode: low values of σ_{yyc} are invariably associated with an interlaminar failure mode for the DCB joint, whilst high values of σ_{yyc} are associated with a cohesive failure through the adhesive layer.

Fractography The fractographic studies on the failed interlaminar DCB specimens revealed the reason why some of the composites possessed relatively low values of σ_{yyc}, which resulted in interlaminar failure of the bonded DCB joints at a relatively low adhesive fracture energy, G_c. Namely, when the value of σ_{yyc} for the composite was low the fracture surface from the interlaminar test revealed bare fibres, see for example, Figure 11. In this micrograph for the unidirectional "Kevlar"-fibre/PA composite the "Kevlar" fibres are clearly visible and appear to have completely debonded from the thermoplastic amorphous polyamide copolymer; the value of σ_{yyc} for this composite was only found to be about 21.5 MPa. In the case of the unidirectional carbon-fibre/PA composite (Figure 12) and the

TABLE VIII

Values of the calculated out-of-plane transverse tensile stresses, σ_{yy}, (from FEA) when DCB joints are loaded to the equivalent of the maximum, plateau values of G_c, the observed locus of joint failure and values of the measured out-of-plane transverse tensile fracture stresses, σ_{yyc}, of the composites

	Data for DCB joints—adhesive employed:				
	Cold-cured epoxy (EA9309.3)		Hot-cured epoxy (FM73M)		
Composite	σ_{yy}	*L* of *F*	σ_{yy}	*L* of *F*	σ_{yyc}
U/d carbon/epoxy	82.0	Il	55.9	Coh	58.9
U/d carbon/PEEK	81.9	Coh	55.8	Coh	84.3
U/d carbon/PA	81.9	Coh	55.9	Coh	83.7
Woven carbon/PA	79.1	Coh	54.2	Coh	83.7
U/d "Kevlar"/PA	71.4	Il	48.8	Il	21.5
Woven "Kevlar"/PA	81.1	Il	54.5	Il	21.5
U/d carbon/PI	82.0	Il	56.0	Il	56.1
Woven carbon/PI	76.5	Il	54.6	Coh	56.1
U/d carbon/PPS	82.0	Il	56.1	Il	29.4

Notes: (i) G_c (plateau) is plateau value of the adhesive fracture energy which for, the thermoplastic composites, occurs after a given corona pretreatment level; obviously this is the uniform constant value for the carbon-fibre/epoxy composite since no corona treatment is needed for this composite to ensure high G_c values and cohesive fracture. (ii) All stresses in MPa. (iii) "Coh": cohesive in adhesive layer failure; "Il": interlaminar fracture in composite.

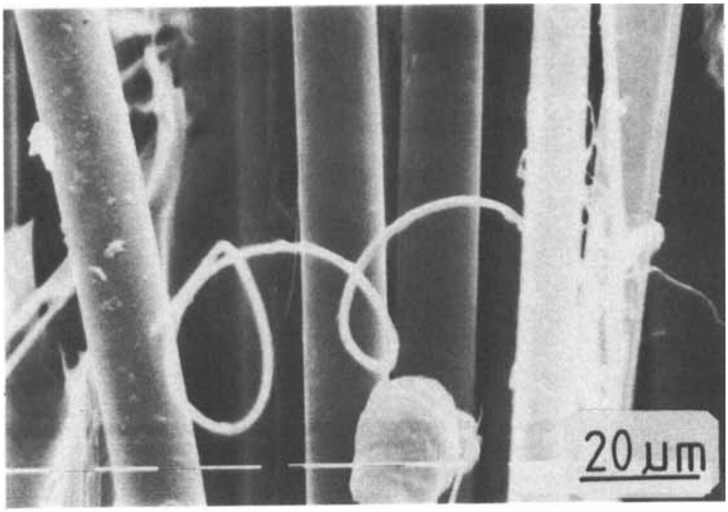

FIGURE 11 Scanning electron micrograph of the failed surface from an interlaminar DCB specimen consisting of unidirectional "Kevlar"/polyamide copolymer composite.

unidirectional carbon-fibre/epoxy composite (Figure 13) for example, the fibres appear to be relatively well embedded in the matrix polymer. Hence, the adhesion across the fibre/matrix interface appears to be higher, which leads to higher values of the transverse tensile fracture stress, σ_{yyc}, and so to less likelihood of the composite substrate delaminating during testing of the bonded DCB joint. Therefore, assuming that the intrinsic adhesion forces across the composite/adhesive interface are sufficiently strong, assured by using a corona

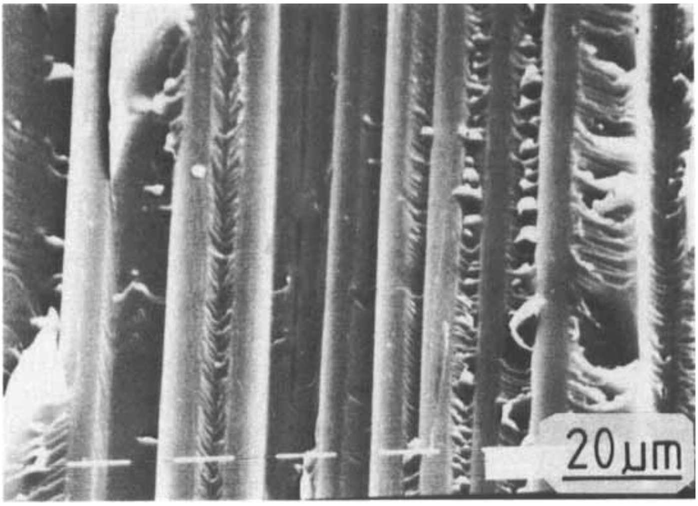

FIGURE 12 Scanning electron micrograph of the failed surface from an interlaminar DCB specimen consisting of unidirectional carbon-fibre/polyamide copolymer composite.

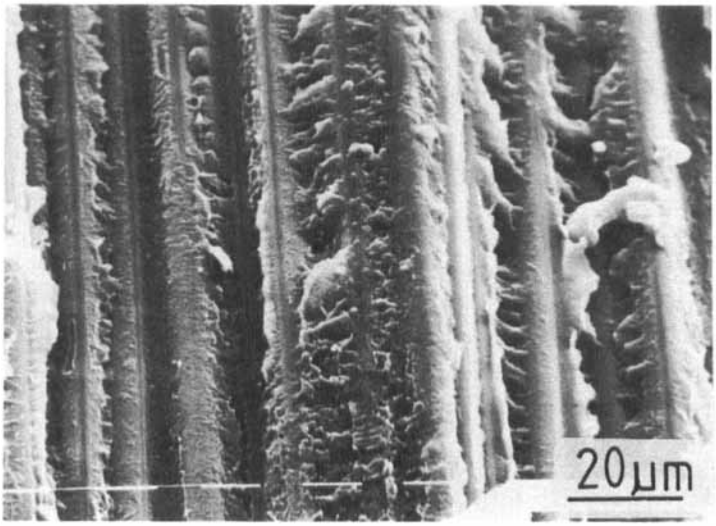

FIGURE 13 Scanning electron micrograph of the failed surface from an interlaminar DCB specimen consisting of unidirectional carbon-fibre/epoxy composite.

pretreatment in the case of the thermoplastic composite, then high values of σ_{yyc} lead to the crack being forced to remain in the adhesive layer and to a relatively high value of the adhesive fracture energy, G_c, being measured.

Stress analysis To extend the above argument to a quantitative description it is necessary to ascertain the stress field surrounding the initial crack tip in the adhesive layer in the DCB joint. This stress field will extend into those regions of the composite substrates above and below the crack tip in the adhesive layer and will give rise to out-of-plane tensile stresses, σ_{yy}, acting to delaminate the composite. If the values of these σ_{yy} stresses can be estimated and compared to the values of σ_{yyc} (i.e. the measured out-of-plane transverse tensile fracture stress of the composite) then the tendency of the composite substrate to delaminate may be quantitatively assessed.

To ascertain the values of the out-of-plane transverse tensile stresses, σ_{yy}, a numerical finite element analysis (FEA) approach was adopted. This consisted of using the "PAFEC" FEA package.[7] For the FEA analysis a DCB joint specimen was directly drawn on a "Sigmax 6164" terminal using the "PIGS"[8] multicolour 2-dimensional package. The terminal was connected to a "VAX" computer on which the "PAFEC" FEA input files were run. Because of the midplane symmetry, only the upper half of the DCB specimen was modelled in the finite element analysis. The experimental situation was simulated on the computer such that the top left hand node of the specimen was point loaded in small loaded increments to avoid sudden deflections and hence errors. This same node was also constrained from moving in the x-direction. The nodes between the crack tip (point A) and the bottom right hand node (point B) were constrained from moving in the y-direction, see Figure 14. The starter crack length was 20 mm. Eight-noded isoparametric-curvilinear-quadrilateral elements were used throughout the specimen, except at the crack tip where four six-noded quarter-point triangular elements were used to account for the singularity of the stresses at the crack tip. The elements were refined near the crack tip region where high stress variations and concentrations were expected. Throughout the analysis 964 elements and 3070 nodes and 1038 elements and 3296 nodes were used for the DCB specimens bonded with the cold-cured epoxy-paste (EA9309.3) (total adhesive thickness 0.4 mm) and the hot-cured epoxy-film (FM73M) (total adhesive thickness 0.5 mm), respectively. The type of elements used were orthotropic for the substrates and isotropic for the adhesives. All the FEA analysis was carried out in a plane-strain condition.

Several assumptions were made for the FEA analysis. Firstly, it was assumed

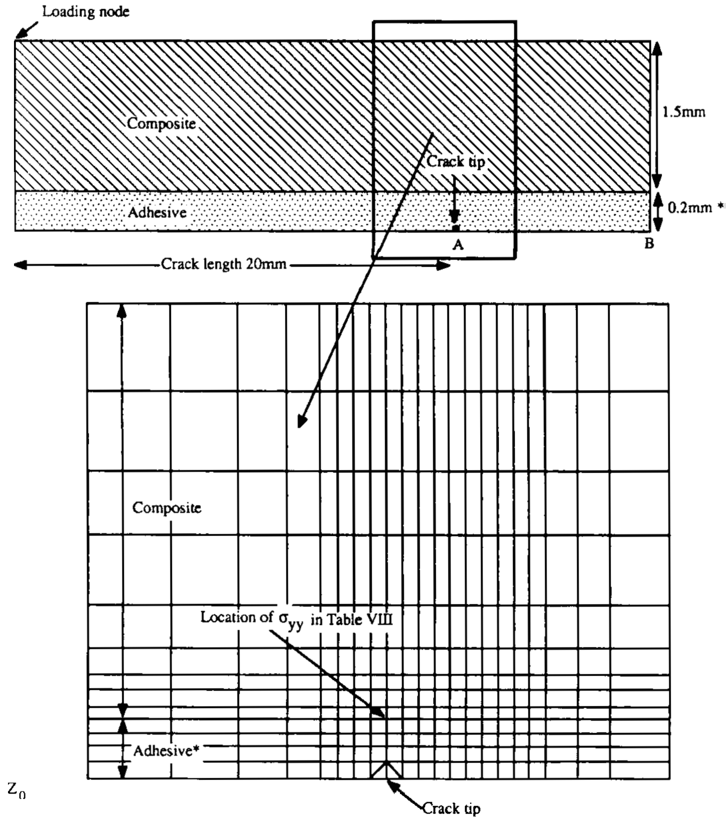

FIGURE 14 The finite element model of the DCB adhesive joint specimen (upper-half) with details of mesh refinement around the crack tip. *Note that for the cold-cured epoxy-paste (EA9309.3) adhesive the thickness of the adhesive layer in the upper half model was 0.2 mm and for this case four layers of elements were used. Whereas, for the hot-cured epoxy-film (FM73M) adhesive the thickness of the adhesive layer in the upper half model was 0.25 mm and for this case five layers of elements were used.

that the adhesives and composites behaved in an elastic manner. Secondly, the maximum loads used in the FEA analysis were calculated *via* Eq. (2), assuming that the crack was going to run in a cohesive manner in the adhesive. For generally orthotropic material analysis "PAFEC"[9] requires the nine directional compliances which are:

$$\begin{array}{lll} S_{xx} = 1/E_{xx} & S_{yy} = 1/E_{yy} & S_{zz} = 1/E_{zz} \\ S_{xy} = -\mu_{xy}/E_{xx} & S_{yz} = -\mu_{yz}/E_{yy} & S_{zx} = -\mu_{zx}/E_{zz} \\ SH_{xy} = 1/G_{xy} & SH_{zx} = 1/G_{zx} & SH_{yz} = 1/G_{yz} \end{array} \tag{3}$$

where S is compliance and SH is shear compliance, μ is Poisson's ratio, E is Young's modulus and G is shear modulus. It should be noted that the fibre reinforcement in this FEA analysis is always in the x–z plane, as shown in Figure 14.

For a transversely isotropic material—an orthotropic material which is isotropic around one axis such as the unidirectional-fibre composites—then there will be only five independent compliances since:

$$S_{yy} = S_{zz} \qquad SH_{zx} = SH_{xy}$$

and (4)

$$S_{xy} = S_{zx} \qquad SH_{yz} = 2(S_{yy} - S_{yz})$$

For a square symmetric material—an orthotropic material which has equal amounts of reinforcement in one plane, in two mutually-perpendicular directions such as woven-fibre composites—then there will be only six independent compliances since:

$$S_{xx} = S_{zz} \qquad SH_{yz} = SH_{xy} \qquad S_{xy} = S_{yz} \tag{5}$$

The material properties used in the FEA analysis for the adhesives and the composites are given in Table IX.

The FEA predictions of the variation of the out-of-plane transverse stress, σ_{yy}, distribution along the y-axis for the DCB joints are given in Figures 15 and 16. These figures show the variation of the transverse tensile stress from the crack tip, which is located in the adhesive, to the adhesive/composite interface and beyond into the composite substrate. The predicted transverse tensile stresses in the adhesive layer agree well with the values of σ_{yy} from the monolithic solution which are given by:

$$\sigma_{yy} = \frac{K_I}{\sqrt{2\pi r}} \cos\frac{\theta}{2}\left[1 + \sin\frac{\theta}{2}\sin\frac{3\theta}{2}\right] \tag{6}$$

where K_I is the fracture toughness of the adhesives, r is the distance from the crack tip and θ is the angle with respect to the x-axis and in this case is 90°. Beyond the adhesive/composite interface the transverse tensile stresses are different from the monolithic solution. This difference can be readily understood, since the composites and adhesives have different material properties. The

TABLE IX
Material property values used in the FEA studies

Material	(a) Adhesive properties Tensile modulus (GPa)	Poisson's ratio
Cold-cured epoxy adhesive (EA9309.3)	1.85	0.35
Hot-cured eopxy film adhesive (FM73M)	2.45	0.35

(b) Composite compliance properties

Material	S_{xx}	S_{yy}	S_{zz}	S_{xy}	S_{yz}	S_{zx}	SH_{xy}	SH_{zx}	SH_{yz}
				($m^2/N \times 10^{-12}$)					
U/d carbon/epoxy	6.67	105	105	−1.73	−31.5	−1.73	200	200	273
U/d carbon/PEEK	7.41	97.1	97.1	−2.15	−29.1	−2.15	208	208	252
U/d carbon/PA	8.0	105	105	−2.32	−31.6	−2.32	200	200	273
Woven carbon/PA	13.7	105	13.7	−3.97	−3.97	−0.82	200	200	200
U/D "Kevlar"/PA	13.1	178	178	−4.41	−53.6	−4.41	476	476	463
Woven "Kevlar"/PA	26.7	178	26.7	−8.0	−8.0	−2.94	145	145	145
U/d carbon/PI	9.1	105	105	−2.7	−31.5	−2.7	200	200	273
Woven carbon/PI	17.5	105	17.5	−5.26	−5.36	−1.4	200	200	200
U/d carbon/PPS	8.13	105	105	−2.44	−31.5	−2.44	200	200	273

Notes: (a) Data for adhesives measured using tensile dumbell specimens at 20°C and at a crosshead displacement-rate of 1 mm/min. (b) Data for composites from manufacturer.

detailed stress variations around the crack tip of composite DCB joint specimens will be given in a separate publication.[4]

Predictions of locus of joint failure The predicted maximum values of the out-of-plane transverse tensile stresses, σ_{yy}, from the finite element analysis at the applied loads on the DCB joint necessary to cause a cohesive fracture through the

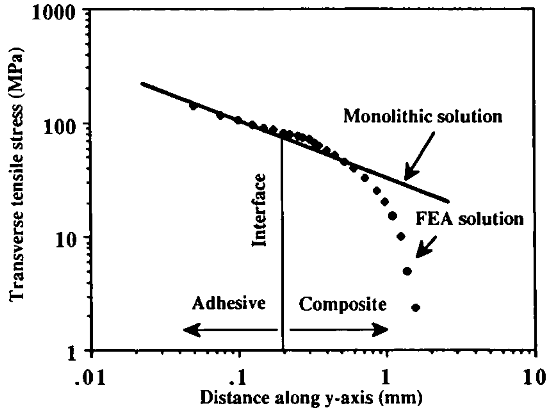

FIGURE 15 The finite element analysis prediction of the out-of-plane transverse stress, σ_{yy}, distribution along the y-axis above the crack tip for unidirectional carbon-fibre/PEEK composite bonded using the cold-cured epoxy-paste adhesive (EA9309.3; total adhesive thickness is 0.4 mm); starter crack length is 20 mm and composite substrate thickness is 1.5 mm.

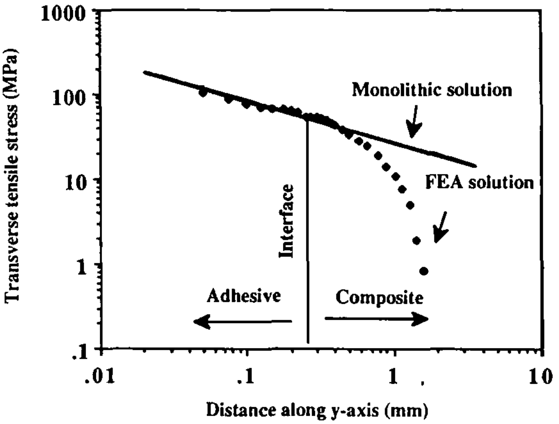

FIGURE 16 The finite element analysis prediction of the out-of-plane transverse stress, σ_{yy}, distribution along the y-axis above the crack tip for woven "Kevlar"-fibre/PA composite bonded using the hot-cured epoxy-film adhesive (FM73M; total adhesive thickness is 0.5 mm); starter crack length is 20 mm and composite substrate thickness is 1.5 mm.

adhesive layer, are given in Table VIII for the various adhesive/composite combinations. Also given in this table are the measured out-of-plane transverse tensile fracture stresses, σ_{yyc}, for the composites. The location of the predicted out-of-plane transverse stresses listed in Table VIII is just inside the composite substrate, above and below the crack tip. Now, for those cases when the calculated values of σ_{yy} are lower than the measured out-of-plane transverse tensile fracture stress, σ_{yyc}, of the composite then it would be predicted that interlaminar fracture of the composite would not occur. Thus, providing the intrinsic strength of the interface is sufficient, the DCB joint would be expected to fail by a cohesive fracture through the adhesive layer. Conversely, when the calculated values of σ_{yy} are higher than the measured out-of-plane transverse tensile fracture stress, σ_{yyc}, of the composite then it would be predicted that delamination of the composite substrate would now be the preferred mode of failure. As may be seen from the data presented in Table VIII, the predictions based upon the above arguments are in excellent agreement with the experimental observations of the locus of failure. Thus, the locus of joint failure for DCB joints is governed by the relative values of the transverse tensile stress, σ_{yy}, in the composite in the region of the crack tip compared to the transverse fracture stress, σ_{yyc}, of the composite; assuming that a sufficiently adequate surface pretreatment has been employed for the composite substrate prior to bonding.

CONCLUSIONS

The main conclusions are:

1. That, as reported in previous publications, for freshly-prepared thermoset-based composites there are generally no problems in attaining adequate adhesion

using epoxy- and acrylic-based structural adhesives; a simple abrasion/solvent wipe pretreatment is sufficient.

2. For the wide range of thermoplastic-based composites which have been examined it is possible to use a hot-melt film adhesive, of the same chemical composition as the matrix, to attain relatively high values of the adhesive fracture energy, G_c, using a simple abrasion/solvent wipe pretreatment for the thermoplastic composites. The values of G_c obtained are very similar to those measured for the interlaminar failure of the composites. However, the use of these films as hot-melt adhesives does, of course, require high temperatures to be reached in the bonding operation.

3. If the epoxy- and acrylic-based structural adhesives are to be employed, then a simple abrasion/solvent wipe pretreatment is now *insufficient* to ensure adequate bonding. This surface pretreatment results in joints possessing very low values of G_c with the locus of joint failure being by crack growth along the weak adhesive/composite interface.

4. A corona-discharge treatment prior to bonding is however, extremely effective in increasing the intrinsic adhesion of the adhesive to the thermoplastic composite, providing the composite is not heavily contaminated with release agent. For the corona-pretreated thermoplastic composite joints the value of G_c increases steadily with the level of corona treatment employed until a maximum, plateau value of G_c is reached.

5. The highest plateau values of the adhesive fracture energy, G_c (plateau), are recorded when the joint fails by crack growth through the adhesive layer. It is of interest to note that under such conditions the value of G_c (plateau) may be far higher than the interlaminar fracture energy, G_c(il), of the composite. However, for some of the joints, before failure *via* crack growth through the adhesive can occur, the thermoplastic composite substrate delaminates. Thus, the composite is now the "weakest link" in the joint and the measured value of G_c (plateau) is now approximately equal in value to the interlaminar fracture energy, G_c(il), of the composite.

6. The tendency for the composite to delaminate in the bonded joint does *not* appear to be governed by the value of the interlaminar fracture energy, G_c(il), of the composite. This tendency is governed by the value of the out-of-plane transverse tensile fracture stress, σ_{yyc}, of the composite relative to the transverse stresses, σ_{yy}, generated in the composite substrate during the DCB joint test. The composites with the lowest values of transverse tensile fracture stress, σ_{yyc}, appear to be those where the fibre/matrix adhesion is relatively poor.

7. The transverse stresses, σ_{yy}, generated in the composite substrates, in the vicinity of the crack tip in the adhesive layer, have been calculated using a finite element analysis approach. By a comparison of the relative values of σ_{yyc} and σ_{yy} the locus of failure observed in the various adhesively-bonded composite may be understood and predicted. This emphasises the importance of not only selecting the appropriate adhesive and surface pretreatment, but also the very significant influence the properties of the composite substrate may have on the measured performance of the bonded composite joint.

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