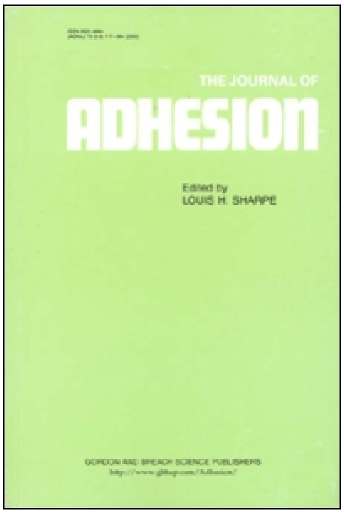


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# The Adhesive Bonding of Thermoplastic Cornposites

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# The Adhesive Bonding of Thermoplastic Composites†

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The present paper reports some initial results on the adhesive bonding of thermoplastic composites, based upon carbon-fibre in a matrix of poly(aryl ether-ether ketone). Both single- and double-overlap joints have been employed and the mechanisms of failure studied using scanning electron microscopy. Further, a theoretical model, based upon a shear-lag analysis, has been used to predict the strength of the double-lap joints as a function of the overlap length and the theoretical results are compared to the experimental data.

## INTRODUCTION

Thermoplastic fibre-composites appear to offer certain major advantages compared to those based upon thermosetting resin matrices.[1] For example, they possess greater toughness, and therefore a higher damage tolerance, and offer the potential of easy and rapid fabrication, *via* thermoforming processes. However, in any component using thermoplastic composites it is likely that adhesive bonding, employing conventional structural adhesives, will be necessary in order to utilise fully the good strength-to-weight ratio of the fibre-composite.

In the case of fibre-composites based upon thermosetting resins, typically epoxy resins, the previous work[2–6] has clearly shown that

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adequate intrinsic adhesion of a structural epoxy adhesive to the surface of the composite may be attained by a surface pretreatment which consists of a simple grit-blast followed by a solvent-wipe to remove any remaining debris, assuming the composite is initially dry and free from excessive release agent.

In the present paper some initial work will be described on the adhesive bonding of a thermoplastic composite based upon carbon fibres in a matrix of the thermoplastic polymer, poly(aryl ether-ether ketone). The aims of this initial study were to determine the strengths of lap joints, identify the mechanims of failure and compare the measured strengths of the double-lap joints, when the out-of-plane tensile stresses (also termed "peel stresses") are relatively low, to theoretical predictions of the failure load based upon a shear-lag analysis.



## EXPERIMENTAL

### The materials

The thermoplastic composite was APC-2 ("aromatic polymer composite 2", supplied by Imperial Chemical Industries, UK) which is a carbon-fibre composite containing a volume fraction of fibres of about 60% based upon a matrix of the thermoplastic polymer, poly(aryl ether-ether ketone). The composite substrate was prepared by laying unidirectional tape into a twelve-ply stack. The ply direction was $[0°, 90°, 90°, 0°, 0°, 90°]_2$; giving a balanced laminate to avoid warping during the moulding process which was carried out in a heated press at 380°C and 1 MPa.

The two commercial adhesives examined were (a) a two-part rubber-toughened epoxy-paste and (b) a one-part modified epoxy-film adhesive.

### Joint preparation and testing

Both single- and double-lap joints were prepared with overlap lengths which varied from 12.7 to 101.6 mm. The composite sheet, 1.44 mm thick, was cut into strips 25.9 mm wide and the surfaces to be bonded were subjected to a light grit-blast and then wiped using a clean cotton cloth dipped in methyl-ethyl ketone. The adhesives were then applied in accordance with the manufacturer's instructions and the adhesive cured. In the case of the two-part epoxy this consisted of subjecting the joints to a temperature of 65°C for sixty minutes under a pressure of 100 kPa, and for the epoxy film adhesive to a temperature of 120°C for thirty minutes, also under 100 kPa. Prior to testing, the thickness of the adhesive layer was measured using a travelling microscope. For the former adhesive the average thickness was 0·09 mm whilst for the latter it was 0.11 mm, with a coefficient of variation of about 10%.

The joints were fractured by loading in tension in an "Instron" tensile testing machine at a crosshead speed of 5 mm/min. When testing the double-lap joints a piece of aluminium, approximately the same thickness as the composite substrate, was inserted between the parallel substrates to avoid distortion of the joint. Five replicates for each type of joint were tested.



### Electron microscopy

The surface of the APC-2 composite prior to bonding and the fracture surfaces of the joints were examined both visually and using the scanning electron microscope. Before observation in the scanning electron microscope the specimens were sputter-coated with gold to improve the conductivity of the surfaces and reduce charging.

### Bulk properties of the adhesive

To ascertain the bulk properties of the adhesive in a state nominally of pure shear, "thick-adherend" shear tests were conducted.[5,7] Aluminium-alloy substrates, 12 mm thick, were employed and were subjected to a chromic-sulphuric etch[8] prior to bonding. Application and curing of the adhesives was conducted as described previously for the composite joints. The thickness of the adhesive layer was carefully measured and a transducer attached to the specimen so as to measure accurately the displacements across the central region of the overlap, which was 12.3 mm long. Joints were tested at a rate of 5 mm/min. So that any deformation in the substrates could be compensated for, a solid aluminium-alloy specimen was also machined and tested and the loads and displacements recorded. Four replicate tests were conducted.

## RESULTS AND DISCUSSIONS

### *Single- and double-lap joint fracture studies*

The results for the failure loads, expressed as force per unit width of joint, for the single- and double-lap joints as a function of overlap length are shown in Table I. Also recorded are the coefficients of variation and the visually assessed locus of joint failure. Several noteworthy features are apparent.

Firstly, for all the joints the failure load increases as the length of the overlap is increased, generally up to a plateau value. This is, of course, a well established observation[4–6,9] and arises from the maximum shear stress concentrations in lap joints being located at the ends of the overlap and being proportional to the overlap length. So once a certain overlap length is attained there is no further gain in failure load as the overlap length is increased.

Secondly, for the shorter overlaps the failure load for the double-lap joint is more than twice that of the single-lap joint with

TABLE I
Failure loads for epoxy/thermoplastic composite joints

| Type of joint | Overlap (mm) | Failure load (MN/m) | C of V | Locus of failure |
|---|---|---|---|---|
| *Two-part epoxy-paste adhesive*: | | | | |
| Single-lap | 12.7 | 0.143 | 12% | I |
| | 38.1 | 0.128 | 22% | I |
| | 63.5 | 0.179 | 23% | I |
| | 102.7 | 0.320 | 11% | I |
| Double-lap | 12.7 | 0.368 | 20% | I |
| | 38.1 | 0.429 | 30% | I |
| | 63.5 | 0.415 | 23% | I |
| | 102.7 | 0.436 | 11% | I |
| *One-part epoxy-film adhesive*: | | | | |
| Single-lap | 12.7 | 0.199 | 36% | I |
| | 38.1 | 0.297 | 29% | I |
| | 63.5 | 0.435 | 21% | I |
| | 102.7 | 0.365 | 22% | I |
| Double-lap | 12.7 | 0.400 | 34% | I; 1 joint some IL |
| | 38.1 | 0.712 | 32% | I; 1 joint some IL |
| | 63.5 | 0.678 | 34% | I |
| | 102.7 | 0.651 | 18% | |

Notes: I = interfacial failure from visual assessment; IL = interlaminar failure in composite substrate.

the same overlap length. This arises from the comparatively high out-of-plane tensile stress concentrations at the ends of the overlap in the single-lap joint and the fact that they decrease in intensity as the overlap lap is increased, *i.e.* as the eccentricity of the loading path and the associated bending moments are reduced.[6,9]

Thirdly, the coefficient of variation in all the sets of joints is relatively high. The joints were carefully made in the laboratory and certainly for the double-lap joints, a coefficient of variation of about 5% to 8%, or so, would be considered more typical of joints with a thermosetting-based carbon-fibre reinforced-plastic.

Fourthly, the locus of joint failure from a visual assessment is that fracture almost exclusively occurred at the adhesive/composite interface, although two joints prepared using the epoxy-film adhesive failed with some degree of interlaminar fracture in the composite substrate. This interfacial locus of failure is most unwelcome to the adhesives technologist and is frequently associated with relatively low joint strengths and a high scatter. Indeed, it is general practice to select pretreatments and design joints to avoid interfacial failure, since it is easier to predict the strength and control the reproducibility when fracture occurs in the adhesive or substrates. In fact, an interfacial locus of failure is especially uncommon when bonding fibre-composites based upon thermosetting resins. Typically, joint fracture either occurs in the adhesive layer or by interlaminar fracture of the composite substrate; although when bonding very thin composite sheets joint fracture may occur *via* tensile failure in the substrates, often well away from the overlap.

Finally, although of interest, it is difficult to compare directly the strength expected if similar joints were prepared from fibre-composites based upon thermosetting resins. However, for the double-lap joints with long overlap lengths, the plateau strength might be expected to be of the order of about 1 MN/m. But since such joints often fail by interlaminar fracture of the composite this may be increased to about 3 MN/m by redesigning the joint at the ends of the overlap to alleviate the out-of-plane tensile stresses that are, of course, present even in a double-lap joint and are responsible for this mode of failure. Typical redesign methods involve tapering the outer substrates, using an angled adhesive spew fillet, etc.[4,6,9] Obviously, since the joints with the thermoplastic composites are apparently failing mainly *via* interfacial fracture, it is doubtful whether such design methods would assist in increasing the joint strength, but this approach is currently being examined.

## Fractography studies

The scanning electron micrograph of the untreated APC-2 thermoplastic composite is shown in Figure 1 and the grit blasted and solvent cleaned composite in Figure 2. As may be seen, the surface pretreatment has roughened the surface considerably and exposed areas of the underlying fibres. Micrographs of the "adhesive" and "composite substrate" from opposite sides of the fracture plane from double-lap joints bonded with the two-part paste epoxy adhesive are shown in Figures 3a and 3b respectively. The composite side of the joint appears to resemble strongly the abraded surface prior to bonding, see Figure 2. There is no sign of any substrate transfer to the adhesive, either from the visual or electron microscopy observations. Thus, the locus of failure does indeed appear to be along the adhesive/thermoplastic-composite interface. Micrographs of the "adhesive" and "composite substrate" from opposite sides of the fracture plane from double-lap joints bonded with the one-part film epoxy adhesive are shown in Figures 4a and 4b, respectively. However, whilst these micrographs suggest that a considerable degree of interfacing failure occurs there is also evidence that some of the outermost layer of thermoplastic matrix

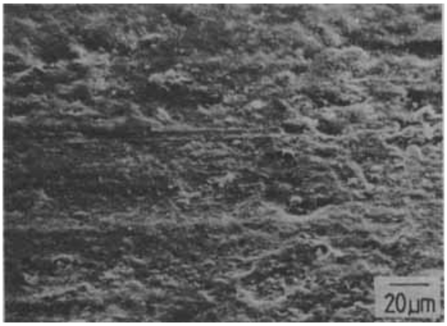
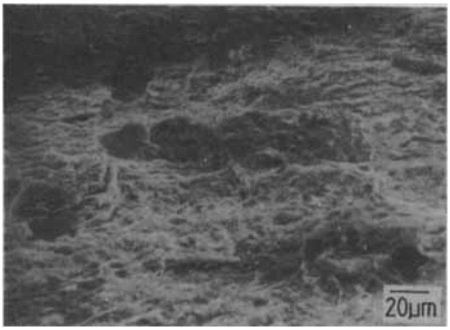
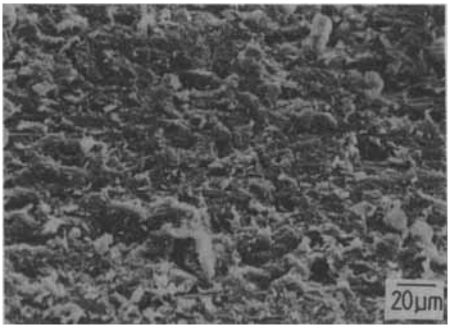



FIGURE 1 Scanning electron micrograph of as-received APC-2 thermoplastic composite.

FIGURE 2 Scanning electron micrograph of abraded/solvent-cleaned APC-2 thermoplastic composite.

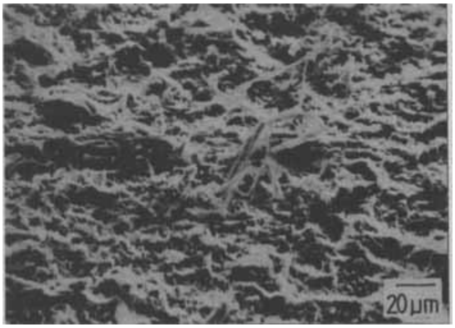


(a)

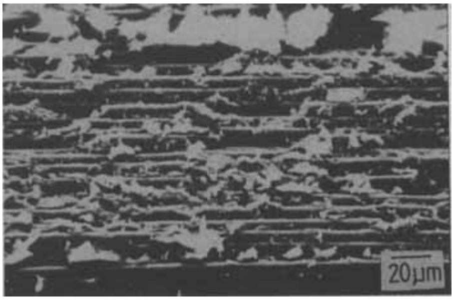


(b)

FIGURE 3 Scanning electron micrograph of fracture surfaces from APC-2/two-part epoxy-paste adhesive joint. (a) "adhesive" side, (b) "composite" side.




(a)




(b)

FIGURE 4 Scanning electron micrograph of fracture surfaces from APC-2/one-part epoxy-film adhesive joint. (a) "adhesive" side, (b) "composite" side.



has failed from the fibres. The adhesive side of the fracture plane (Figure 4a) appears to contain some highly-drawn fibrous structure which is not typical of crosslinked epoxy adhesives but might be the thermoplastic matrix of the composite. Also, the composite side (Figure 4b) shows far more exposed fibres compared to the original abraded surface (Figure 2), confirming transfer of some of the outer layer of matrix. Thus, for the joints bonded with the epoxy-film adhesive there is a mixture of failure modes, namely: interfacial, matrix/fibre and interlaminar, the last being observed visually on two joints, as recorded in Table I. The lesser extent of interfacial failure with this adhesive may obviously well be responsible for the higher joint strengths which were observed (Table I).

**Comparison of theoretical and experimental joint strengths**

One aim of the present initial work was to try and predict the failure loads of the joints using various theoretical analyses. However, all the analyses are based upon the assumption that the adhesive or substrate will be the weakest part of the joint and one cannot predict accurately the joint strength when interfacial failure occurs. The types of joint where least interfacial failure is observed are the double-lap joints employing the epoxy-film adhesive. It is therefore of interest to compare the theoretical and experimental data for this type of joint.

The three failure modes that may be considered theoretically are (a) tensile fracture in the adhesive or "bulk" interlaminar fracture in the composite substrate, (b) shear failure of the adhesive and (c) tensile fracture in the composite substrate, well away from the bonded overlap. The data used in the various analyses are shown in Table II.

Considering firstly the possibility of tensile fracture in the adhesive or "bulk" interlaminar fracture in the composite substrate, then these failure modes arise from the presence of the out-of-plane tensile stresses, $\sigma_{11}$. For an elastic adhesive they may be calculated from the equation.[6]

$$\frac{\sigma_{11}(\max)}{\tau_{af}} = \left[\frac{3(1 - v_s^2)E_a}{E_s}\right]^{1/4} \left(\frac{d_0}{h_a}\right)^{1/4} \tag{1}$$

where many of the parameters are defined in Table II, $d_0$ and $h_a$ are the thickness of the outer substrate and adhesive layer respectively and $v_s$ is the Poisson's ratio of the substrate. Equation (1) yields a

TABLE II
Properties of the epoxy-film adhesive and the substrates used in the theoretical analysis

| | |
|---|---|
| *Adhesive properties*: | |
| Shear modulus | 0.80 GPa |
| Young's (or tensile modulus), $E_a$ | 2.16 GPa |
| Maximum elastic shear stress | 31.5 MPa |
| Plastic shear stress at fracture, $\tau_{af}$ | 38.8 MPa |
| Plastic shear strain at fracture | 23 % |
| *Thermoplastic-composite substrate properties*: | |
| Young's (tensile) modulus, $E_s$ | 64.1 GPa |

value of $\sigma_{11}$(max) of about 40 MPa. This is well below the expected tensile fracture stress of the adhesive (about[9,10] 55 to 75 MPa) and the "bulk" transverse fracture stress of the thermoplastic (about[11] 60 MPa). Hence, the theoretical predictions suggest that tensile fracture in the adhesive or "bulk" interlaminar fracture in the composite substrate is unlikely.

Secondly, considering the fracture strength when the joint fails *via* a shear failure in the adhesive, then the theoretical analysis used was based upon a shear-lag analysis[12] and a computer program, to run on an Apple Macintosh, was written to solve numerically the equations relating the loads to the induced displacements in the joint. The substrates are treated as elastic materials but the adhesive is modelled as an elastic-plastic material. The failure criterion assumed is that joint failure occurs when the shear strain in the adhesive layer, at the ends of the overlap length, attains the value of the maximum shear strain capability of the adhesive. This latter property, along with other engineering shear-properties of the adhesive, was measured using the "thick adherend" test. This test

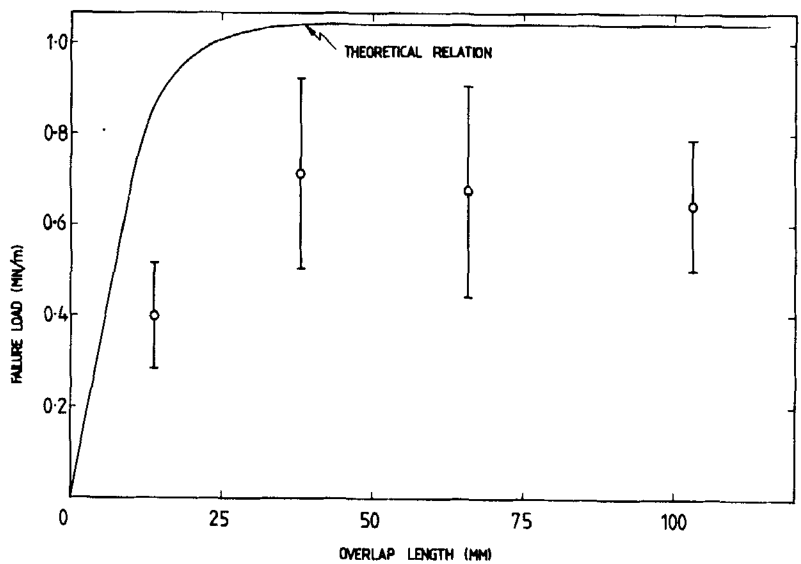



FIGURE 5 Experimental and theoretical relationships between failure loads and overlap length for double-lap joints consisting of APC-2 bonded with an epoxy adhesive.

method was found to give very reproducible results and the data used in the analysis are shown in Table II.

The results of the theoretical analysis are shown in Figure 5, where they are also compared to the experimental results. Both have the same form of relationship between the failure load of the joint and the overlap length. However, the theoretical curve is well above the experimentally measured strengths, again possibly reflecting the considerable amount of interfacial failure observed in these joints.

Thirdly, it is of interest to note that if the joint failed in the composite substrate, well away from the bonded overlap, then the joint strength would be about 1.2 MN/m. This obviously represents the maximum value that may be achieved and values of this order should be obtainable if adequate intrinsic adhesion at the interface(s) is established and attention is given to the details of the joint design.



## CONCLUSIONS

The adhesive bonding of a carbon-fibre thermoplastic composite (APC-2) using conventional structural adhesives appears to present some new problems compared to bonding composites based upon thermosetting resin matrices.

The main difference is that joints using the thermoplastic composites exhibit a high degree of interfacial failure. Further, in the case of the better adhesive there is evidence that the joint also fails by the outermost region of the thermoplastic matrix becoming debonded from the fibres. On the other hand, for thermosetting-composite joints a similar abrasion pretreatment to that used on the thermoplastic composites substrates is extremely effective and interfacial failure is rarely observed.

The observed failure mode resulted in the thermoplastic-composite joints possessing relatively low joint strengths and the associated coefficient of variation being high. Further, with such an interfacial failure mode it is difficult to design and predict the failure behaviour of joints. Indeed, whilst a theoretical shear-lag analysis resulted in the correct form for the strength *versus* overlap relation, the predicted strengths, assuming fracture in the elastic-plastic adhesive layer, were significantly higher than the best of the experimentally measured strengths.

Obviously more work is needed to completely identify the mechanisms of failure in thermoplastic-composite joints but it appears that interest should be focussed upon the level of intrinsic adhesion at the adhesive/composite and matrix/composite interfaces. Such studies are currently in progress.

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