Fiber Orientation Effects in Supported Adhesives

The use of fabric supports (carriers) in structural film adhesives has been established for some time. Such carriers are commonly glass, nylon, or polyester in woven, mat, or knitted form. Supported adhesives may be regarded as fiber-reinforced composites of low fiber volume fraction. In the case of a woven glass carrier, where the fiber is of much higher modulus than the matrix and the fibers are aligned in two directions, anisotropy of properties is a possibility, and some evidence of this in the dynamic mechanical properties of an adhesive has been observed.[1] Although the effect of carriers on such properties as fatigue strength and durability of bonded joints has been examined to some extent,[2-4] the effect of fiber orientation in adhesives containing relatively high modulus carriers does not appear to have been reported. Some observations on this topic are presented here.

EXPERIMENTAL

Two adhesive systems were used: (1) a commercially available, 177°C curing, glass-fiber-supported, epoxy adhesive for aerospace applications (overall composition shown in Table I) and (2) cast films of the Shell epoxy resin Epikote 828 and methylene dianiline (MDA) in the proportion 100:29, containing either (a) the same glass carrier as in (1) obtained by extraction from the uncured adhesive, (b) a polyester mat, or (c) no carrier.

The glass fabric, approximately 32.8 g/m^2, of an open, plain weave, constituted about 5% by volume in the commercial adhesive, as determined by ashing.[5] The polyester mat was approximately 8.0 g/m^2; four layers of this were incorporated in the cast films. The construction of the two fabrics is shown in Figure 1.

Dynamic mechanical properties were measured with a Rheovibron Model DDV-II-C dynamic viscoelastometer operating at 110 Hz and a heating rate of about 2°C/min. Samples were prepared in the following manner: in the case of the commercial adhesive, specimens were cut from the uncured film in three orientations, namely, parallel to the two fiber directions and at 45° to the fiber directions (designated orientations A, B, and C; see Fig. 1). The three specimens were cured simultaneously, using a polyester or PTFE template, in a heated platen press for 1 h at 177°C and 0.3 MPa. The 828/MDA sheets, approximately 1200 × 1200 × 0.3 mm were cast between glass plates coated with a release agent and cured for 45 min at 60°C followed by 30 min at 80°C and then 2.5 h at 150°C. Samples with the desired fiber orientation were cut from the cured sheets. Rheovibron samples were about 50 × 5 mm in all cases.

Joints bonded with the commercial adhesive were tested in tensile-shear (Al–Al single lap joints[6]) and flexure (four-point bending of honeycomb sandwich panels[7] and three-point bending of Al–Al laminates[8,9]). In each case joints were made with the fabric in the three orientations, as above, using 2024-T3 Alclad aluminum and 5052, 0.32 mm cell size aluminum honeycomb. Surface preparation for the aluminum sheet was a vapor degrease in 1,1,1,-trichloroethane followed by a chromic acid etch. Joints were bonded in a heated platen press using a cure cycle of 1 h at 177°C and 0.3 MPa.

TABLE I
Overall Composition of Film Adhesive

Component	Amount (wt %)
Triglycidyl(4-aminophenol)	33
Cresol novolac epoxy resin	30
Diglycidyl ether of bisphenol A	11
Dicyandiamide	8
Asbestos	8
Woven glass support	10

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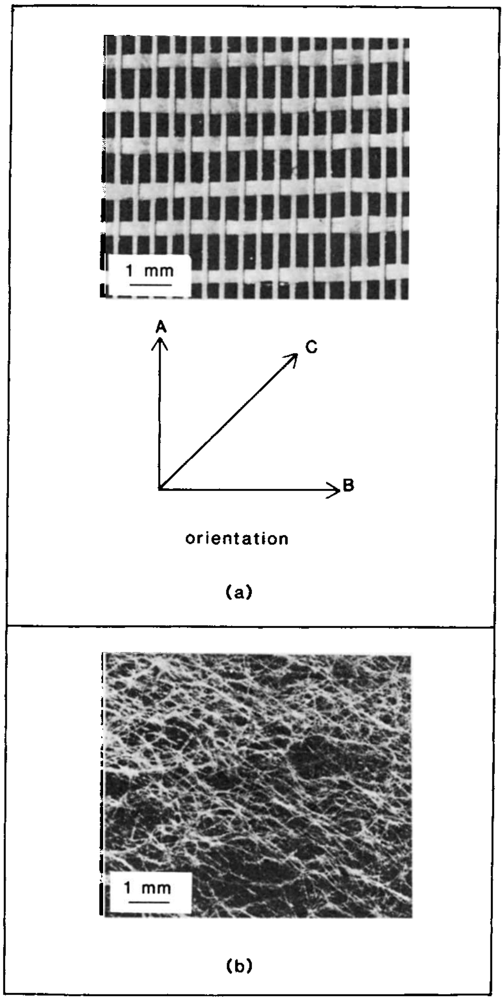

Fig. 1. (a) Woven glass support, (b) polyester mat.

RESULTS AND DISCUSSION

As depicted in Figure 1, the spacing of the fibers of the woven glass support differs in the warp and weft directions, and the fiber roving is wider in one direction than in the other, although the number of ends per roving is the same.[10] Accordingly, samples for dynamic mechanical analysis contained more fiber in the longitudinal direction (parallel to the force axis) for the A orientation than for the B, while samples with the C orientation had no fibers parallel to the force axis. The curves for tan δ and the dynamic modulus E' for the commercial adhesive in the three orientations are shown in Figure 2. A substantial effect of fiber orientation is evident. Similar tests on another commercial film adhesive, on a woven nylon 6,6 support, showed no such effect.[1]

Figures 3 and 4 show the tan δ and dynamic modulus curves for the 828/MDA systems. It is seen that the curves for unsupported material and those supported on the polyester mat and glass fabric in orientation C are similar (apart from the indication in the tan δ curve of the T_g of the polyester at about 110°C). Glass fabric in the A and B orientations clearly makes the samples significantly stiffer over the whole temperature range.

In a study of a single ply of a carbon-fiber-reinforced epoxy composite,[11] it was noted that the height of the α-transition peak was somewhat reduced by fiber at 30° to the stress direction,

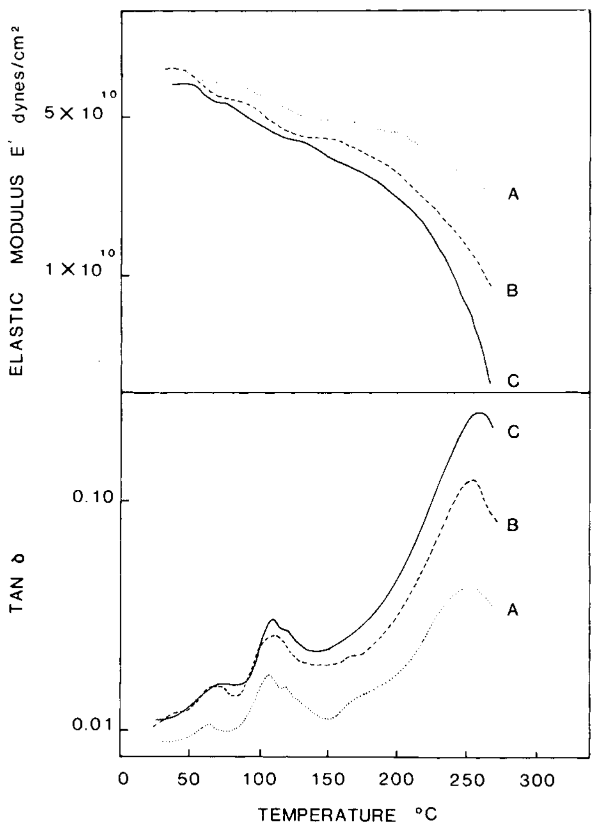

Fig. 2. Dynamic mechanical analysis of a commercial adhesive on a woven glass support. Letters denote fiber orientation (see Fig. 1).

compared to the values for the epoxy resin alone. The β- and ω-transitions (at -50 and 100°C, respectively, for that epoxy resin) were virtually masked by the presence of the fiber. In the present study, no evidence was seen of masking of the transitions at temperatures below the α-transition, but the fiber volume fraction in this case would be much less than in the carbon fiber composite.

The effect on dynamic mechanical properties, of the unsupported 828/MDA system, arising from modification of the network structure, has been studied.[12] The changes in tan δ values over a wide temperature range and in the position of the loss peak were interpreted in terms of the different molecular weight between crosslinks, arising from variations in the 828 to MDA ratio. In the present case, matrix stoichiometry is constant and the tan δ values are virtually in the same order over the whole temperature range, (apart from the influence of the T_g of the polyester support, noted above). Corresponding effects are shown in the dynamic modulus curves (Fig. 4). Evidently, the matrix is reinforced over the whole temperature range, implying good adhesion between matrix and support in each case. The constancy of peak temperature indicates minimal interference in the epoxy cure mechanism by the presence of the fibers.

Using the commercial film adhesive, the question of whether the observed effects of fiber orientation on the dynamic mechanical properties are translated into a modification of adhesive performance was examined by measurements of tensile-shear strength of Al–Al single lap joints and flexure tests on honeycomb sandwich panels and Al–Al laminates. The tensile-shear results are shown in Table II. Four-point flexure tests on honeycomb sandwich panels, tested at 20°C, gave the same maximum force value for each orientation, 7.95 ± 0.05 kN, which was the force to crush the honeycomb. At this value, no delamination was observed.

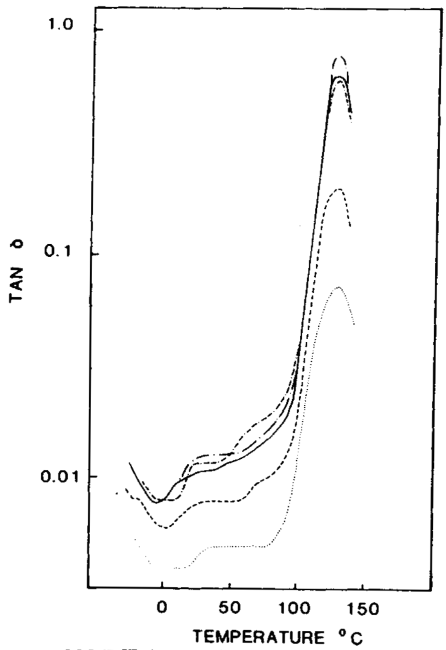

Fig. 3. Tan δ curves for cast 828/MDA samples containing various supports: (- · -) no support; (- - · · - -) polyester mat; (——) C glass; (- - -) B glass; (· · ·) A glass.

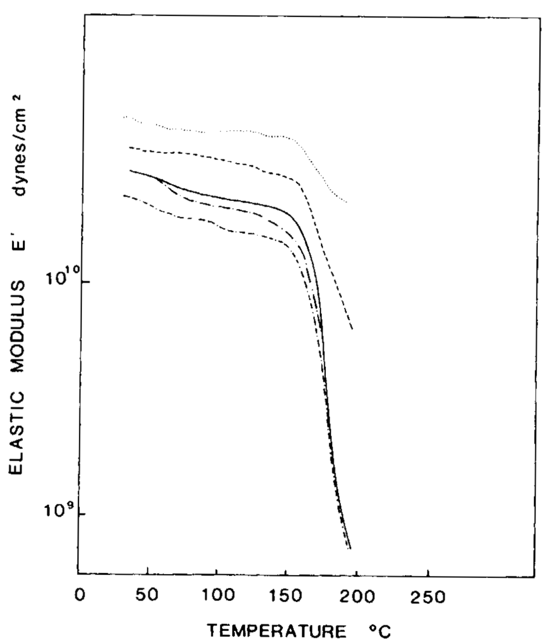

Fig. 4. Dynamic modulus curves for cast 828/MDA samples containing various supports: (- · -) no support; (- - · · - -) polyester mat; (——) C glass; (- - -) B glass; (· · ·) A glass.

TABLE II
Effect of Fiber Orientation on Tensile-Shear Strength of Al–Al Single Lap Joints

Fiber orientation[a]	Tensile-shear strength [MPa (SD)] Test temperature 20°C	177°C
A	20.48 (0.99)	18.15 (0.56)
B	18.66 (0.70)	18.95 (0.57)
C	20.59 (1.41)	17.07 (0.58)

[a] Orientation with respect to the axis of the applied force, i.e, A parallel to force axis.

Results of the three-point flexure test on specially prepared Al–Al laminates (see Fig. 5) are given in Table II.

Figure 2 indicates that the range in modulus values for the three orientations is about 5% at 30°C and about 70% at 180°C. Consequently, it would be expected that any orientation effects on joint strength would be most evident at high temperatures. The tensile-shear results in Table II suggest that the joints with the glass fiber support in the C (diagonal) direction may be less strong at 177°C than the other orientations. Roche et al.[8,9] indicated that the slope of the force/deflection curve of their three-point flexure test is proportional to the modulus of rigidity. In our tests, the decreasing order of the slopes was found to be A, B, C, the same as the order of the dynamic modulus values. The order of the various force values in Table III is, in each instance, A, C, B. The amount of glass fiber in the A direction is approximately twice that in the B direction (Fig. 1). It appears that in the bending mode orientation C is equivalent to the resultant of the A and B configurations.

CONCLUSIONS

Dynamic mechanical analysis of epoxy-based adhesives is substantially affected by the orientation of high modulus fiber supports, such as glass, even though the volume fraction of support is small, but unaffected by supports of modulus similar to the epoxy matrix. For certain joint configurations, there is some suggestion of an effect on adhesive joint performance at those temperatures where the modulus dependence on fiber orientation is greatest.

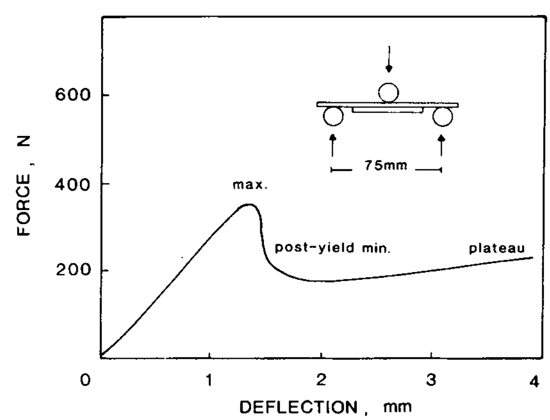

Fig. 5. Three point flexure test configuration and representative force–deflection curve.

TABLE III
Effect of Fiber Orientation on Flexural Strength[a] of Al–Al Laminates

Fiber orientation[b]	Max. force[c] [N (SD)]	Post-yield min. force[c] [N (SD)]	Plateau force[c] [N (SD)]	Initial slope[c] [N/mm (SD)]
A	415 (8)	222 (17)	267 (7)	318 (2)
B	360 (9)	152 (6)	245 (8)	308 (7)
C	392 (11)	200 (6)	252 (3)	303 (7)

[a] Test temperature 177°C.
[b] Orientation with respect to long axis of joint.
[c] See Figure 5.

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