Effects of Surface Preparation on Long-Term Durability of Composite Adhesive Bonds

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ABSTRACT

The determination of the long-term effects of surface preparation techniques for composite bonded joints is addressed. Several potential factors are evaluated by concentrating on the effects of peel plies and grit blasting on the strength and failure modes of adhesively bonded composites. Initially, an evaluation of the floating roller peel test configuration is described, where the intent was to extract quantitative data from this commonly used quality control test method. Subsequently, a unique form of the double cantilever beam (DCB) test was developed and used for a sequence of test evaluations. The research is to aid the interpretation of a form of the well-known wedge crack test where the usual aluminum adherends are substituted by composite adherends.

DCB tests have shown that nylon peel ply surfaces tend to precipitate interfacial failures and intermittent crack propagation, with reduced loads and crack opening displacements, hence significantly lower critical strain energy release rates (G_{Ic}) than equivalent PTFE vacuum bag surfaces. Additionally, grit-blasted bonded joints tend to have higher failure load and G_{Ic} values than non-blasted ones, though the mode of failure (interfacial or cohesive) is unchanged. Several improvements to specimen preparation and testing, including a custom bonding jig and bondline thickness control methods, are also described.

INTRODUCTION

A common practice currently adopted for general aviation aircraft structural development is the use of full-scale structural articles. Unfortunately, this approach can limit the ability to evaluate the adequacy of critical structural details such as bonded joints. By implementing an appropriately-tailored version of the building block test/analysis/fabrication philosophy that supports effective integrated product development and is used widely in the military and commercial aircraft community, valuable and key information can be obtained on bonded joint characteristics. It is suggested that the utilization of such approaches could serve to complement full-

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scale test information, providing general aviation aircraft developers with insight that would facilitate the design and manufacture of reliable bonded structures.

Advantages of bonding over mechanical means of fastening include higher stiffness, more uniform load distribution, cleaner lines for aerodynamic benefits, part consolidation, no holes drilled in adherends (with the resulting stress concentrations), and, generally, less labor.

Adherend surface preparation plays a critical role in developing bonded joints. General aviation tends to rely more extensively on bonded joints, in part due to the lower load intensities typically found in smaller aircraft. Inadequate surface roughening, environmental effects, peel ply chemical contamination [1], and other factors (both mechanical and chemical) can prevent adhesives from bonding properly to composites, resulting in interfacial failures. These failures occur at loads well below those of properly bonded joints that fail cohesively. Other failures can occur over time in service, as joints are exposed to harsh environments, including elevated temperature and humidity [2-11].

Specimens are evaluated with modified versions of the following tests: ASTM D3167 Standard Test Method for Floating Roller Peel Resistance of Adhesives, ASTM D3433 Standard Test Method for Fracture Strength in Cleavage of Adhesives in Bonded Joints, and ASTM D3762 Standard Test Method for Adhesive-Bonded Surface Durability of Aluminum (Wedge Test). Analytical and numerical models of test methods are performed to analyze test data and specimen configuration. Materials and processes typical of those used for aircraft are studied to quantify the relative importance of each factor's contribution to bond strength. Results can be used to provide manufacturers with bonding guidance and to assist the FAA with interpreting data related to certification and evaluation procedures.

LONG-TERM ENVIRONMENTAL VARIABLES AND PHENOMENA

Initially, many possible factors that could affect an adhesive bond's durability were amassed and evaluated (Table I). In this paper, the focus is primarily on the effects of peel plies and grit blasting, both of which affect bonds greatly and are relevant to the aviation industry.

TABLE I. POTENTIAL BONDNG FACTORS

Factor	Variables
Adherend Layup	$0°_{[n]}$, quasi-isotropic, other layup; orientation of ply on bonding surface
Adherend Material	Fiber, matrix, metal, aviation materials
Adhesive Filler Material	Type of filler, percentage of filler
Adhesive Preparation	Hand-mixed, machine-mixed, apply vacuum to remove trapped air
Bondline Thickness Control	Glass microbeads/silane treatment, wires, tabs/tape, applied pressure
Compressed "Shop Air" Blowing	Pressure, exposure time
Grit Blast	Pressure, grit size, number of passes, speed of passes
Hand Sanding	Grit size, number of passes, pressure applied
Humidity Exposure	Humidity %, exposure time, pre-bond, post-bond, under load
Peel Ply	Nylon, polyester, none
Solvent Wiping	Acetone, isopropyl alcohol, number of wipes, applicator type
Temperature Exposure	Temperature, exposure time, pre-bond, post-bond, under load
Water Bath	Temperature, exposure time, pre-bond, post-bond, under load

FLOATING ROLLER TEST REVIEW

The initial approach utilized floating roller tests on woven fiberglass and carbon fiber/epoxy samples. The ASTM D3167 test is designed for a thick adherend bonded to a thin metal adherend that bends around the roller during peeling. Because composite-to-composite bonds are more typical of the type of aircraft being studied, single woven plies were used for the thin adherend instead of metal. However, these thin composite adherends did not conform to the fixture's roller as they were being pulled from the thick adherends. They bent at too tight a radius of curvature and fractured before the bond could be broken (Figure 1). This test method was abandoned in favor of other bond strength tests that do not require such extreme strains on the adherends to fracture the bond.

DCB / WEDGE TEST

Literature research and a review of standard test methods revealed that the DCB and wedge tests (Figure 2) are well-suited to evaluating the short- and long-term durability adhesive bonds [6, 12-17]. In the DCB test, a bonded sample is pulled apart, at a constant test machine crosshead velocity, by fixtures (hinges or pinned blocks) at the end of the beams. The specimen is loaded and unloaded until the crack has propagated entirely through the sample. The wedge test can be performed with the same specimen, but an angled wedge is driven into the crack opening to stimulate crack growth. Then, the sample is observed (often in an environmental exposure chamber) and the crack tip propagation is recorded.

DCB Test Specimen Geometry

Unfortunately, there is a lack of standard test procedures for measuring the bond strength of adhesively bonded composite materials. ASTM test methods researched

Figure 1. Typical versus ideal floating roller test.

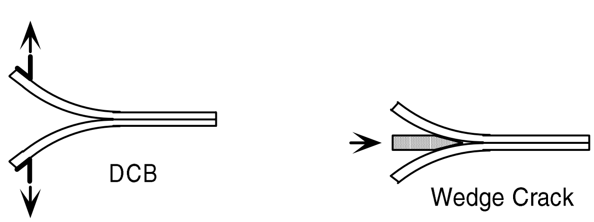


Figure 2. DCB and wedge crack tests.

cover either adhesively bonded metals or interlaminar failures in composites. The new specimen used for the DCB and wedge crack tests is based on those of ASTM D3433, ASTM D3762, and ASTM D5528-94a Standard Test Method for Mode I Interlaminar Fracture Toughness of Unidirectional Fiber-Reinforced Polymer Matrix Composites (Figure 3).

DCB Test Specimens

Two series of DCB tests were performed on IM7/8552 22-ply unidirectional adherends bonded by Dexter Hysol EA9394 two-part epoxy adhesive mixed with 0.005"-diameter glass microbeads (2.5% by weight). All adherends were cured with Chemfab VB-3 PTFE vacuum bag (VB) film on the bottom surface (tool side) and with a nylon peel ply (PP) on the top side. This layup process creates panels with different surface properties on each side. Samples were bonded in one of three different orientations: PP to PP, VB to VB, or PP to VB, with half of each group of samples grit-blasted before bonding, creating six different types of specimens. Although this sort of layup and bagging procedure, with different surfaces on both faces, is not typical of a component used in production, it was better suited as a research aid. In doing so, bonds made to different surface types can be compared against each other more reliably, as all of the specimens are derived from the same panel, removing variances in specimen production.

The composite samples that were grit-blasted (with Mil-A-2222B grit) were done so at 40 psi (regulator line pressure) in the first set and 60 psi in the second set. The hinges used to hold the samples in the test machine grips were made from 0.04" thick continuous hinge, cut to 1" lengths and grit-blasted at 80-100 psi. The surfaces to which the hinges were bonded on all samples were also blasted at 40-60 psi prior to bonding. The hinges were bonded to the samples with the same adhesive and bonding process used for the samples themselves. All bonded surfaces (adherends and hinges) were cleaned thoroughly with de-ionized water, oven-dried, wiped with isopropyl alcohol, then air-dried before bonding. All crack initiators were created with 3" of flashbreaker tape on the ends of the specimens.

Before testing, the sides of samples were spray painted white and 1/16" tick marks were penciled on manually for visual crack tip observation during the tests.

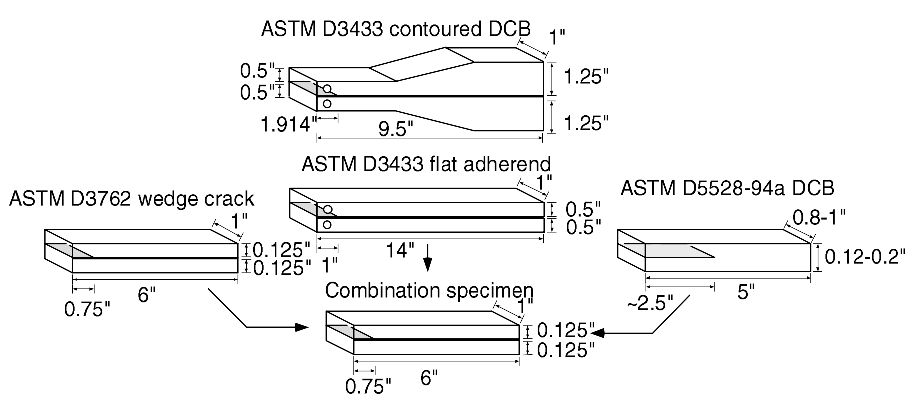


Figure 3. ASTM test specimens combined into one sample geometry.

DCB Test Method

Specimens were loaded into an Instron 8562 test machine by clamping the hinges in test grips. The test machine ran under displacement control at a crosshead speed of 0.02 in/min while loading the sample and at a higher rate while unloading. After visible crack propagation, the sample was unloaded and then reloaded. This process repeated for each ½" of crack growth. Calculations of G_{Ic} were obtained by using Equation 1 from Whitney and Browning [18], which utilizes the area method,

$$G_{Ic} = \frac{1}{2b\Delta a}\left(P_1\delta_2 - P_2\delta_1\right) \tag{1}$$

where P_1 and P_2 are the applied loads at times 1 and 2, δ_1 and δ_2 are the deflections at times 1 and 2, and Δa is the crack length change from time 1 to time 2. The test machine recorded load and opening displacement (at the free ends of the cantilever beams) while the operator noted the crack tip location visually with a monoscope.

DCB Test Results

From a sampling of four typical load-displacement curves, one from each major category (Figure 4), several clear trends emerged:

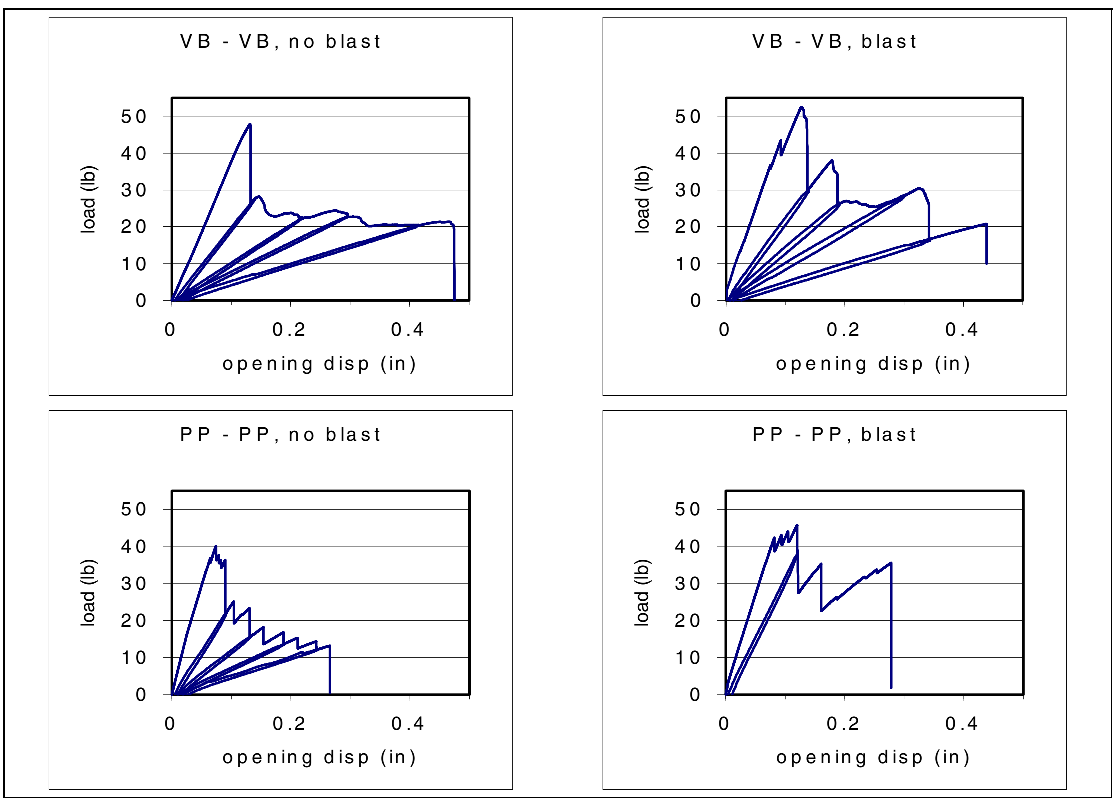


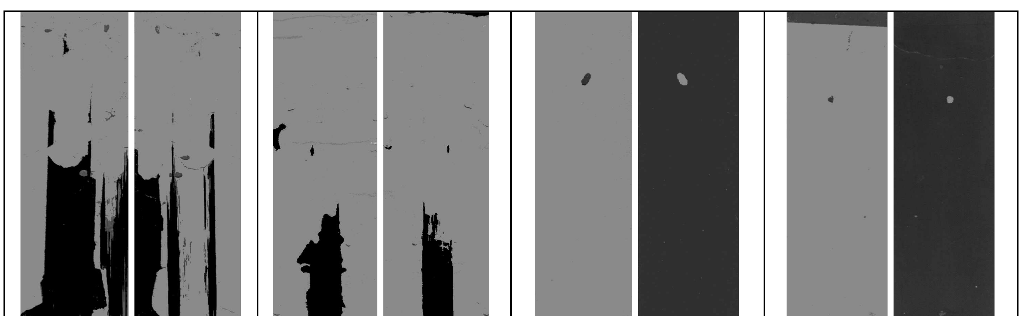
Figure 4. Sample load-displacement curves for bonded composite DCB tests.

1. Bonds made to nylon peel ply surfaces held less load than bonds to vacuum bag surfaces.
2. Bonds made to nylon peel ply surfaces failed at lower opening displacements than bonds to vacuum bag surfaces.
3. Cracks propagated less smoothly in bonds made to nylon peel ply surfaces than in bonds to vacuum bag surfaces.
4. Grit blasting resulted in an increase in the initial failure load.

Just as important as quantitative values like loads, displacements, or energy release rates is the more qualitative analysis of modes of failure. Well-bonded joints should fail within the adhesive (cohesive failure) or within the adherends (interlaminar failure) when broken apart. Failure at the adherend-adhesive interface (interfacial failure) generally indicates that the bond was not performed properly. From a sampling of computer-enhanced (to better distinguish the dark gray adhesive from the black adherends) scans of four typical failure surfaces, one from each of the four main groups of the second set of samples, (Figure 5), a few more trends were clear:

1. Bonds made to nylon peel ply surfaces failed interfacially.
2. Bonds made to vacuum bag surfaces failed cohesively and interlaminarly.
3. Grit blasting surfaces did not change the mode of failure.

The critical strain energy release rates calculated for the DCB test specimens (Table II) follow naturally from the observed trends in the load-displacement curves (Figure 4) and the fracture surfaces (Figure 5). Dexter documents a mode I critical strain energy release rate G_{Ic}=5.83 in-lb/in^2 for their EA9394 adhesive (in their technical service laboratory report), tested on phosphoric acid anodized and etched aluminum with a 0.005" bondline controlled by glass beads. The two key G_{Ic} trends seen in UCSB's tests were:



Bond	VB – VB, no blast	VB – VB, blast	PP – PP, no blast	PP – PP, blast
Failure	Cohesive/interlaminar	Cohesive/interlaminar	Interfacial	Interfacial

Figure 5. Sample computer-enhanced fracture surfaces of bonded composite DCB specimens. Gray areas are adhesive; black areas are adherend.

TABLE II. DCB CRITICAL STRAIN ENERGY RELEASE RATE TEST RESULTS

	PP - PP, No blast	**PP - PP, blast**	**PP - VB, no blast**	**PP - VB, blast**	**VB - VB, no blast**	**VB – VB, blast**
G_{Ic} (in-lb/in^2) avg values	1.319	2.335	N/A	3.769	2.354	2.774

1. Bonds made to vacuum bag surfaces produced higher G_{Ic} values than bonds to nylon peel ply surfaces.
2. Bonds made to grit-blasted surfaces produced higher G_{Ic} values than their non-blasted counterparts, regardless of previous surface preparation.

DEVELOPMENT OF ENHANCED FABRICATION AND TESTING PROCEDURES

Several upgrades to the test method are under consideration for future DCB tests, including an alternate method of crack growth monitoring, discussed below. The specimen preparation itself is undergoing refinement to ensure consistency in bonds from sample to sample, in order to validate comparisons. Additionally, the use of glass microbeads for bondline thickness control is being evaluated.

The DCB tests are intended to provide a foundation upon which to extend the investigation of future wedge testing on similar geometrical configurations. The samples themselves will be the same as those used in the DCB test (minus the hinge hardware) to make data correlation between the two tests straightforward, as they are essentially displacement control vs. load control of the same test (Figure 2). In the wedge test, the specimens are subjected to elevated temperature and humidity, which is a reliable short-term method to predict bond integrity of a joint over long periods of time in service, as detailed in the literature [3-11].

Test Method Refinement

Visual tracking with a monoscope is one method of measuring crack tip location. Other means include the use of a video camera and monitor or the application of crack gauges. Crack gauges can be bonded onto the side of the sample, covering the area where the DCB crack will grow. As the crack opens up the sample, it also creates a slit in the crack gauge, changing the electrical resistance between its leads. A calibrated machine measures the resistance and transforms it into a precise crack length measurement. Unfortunately, typical crack gauges long enough for DCB samples cost $35-$50 apiece, while long, thin DCB gauges are priced at $180 each, making their cost too prohibitive for a large series of tests. They would be better suited to be used in a smaller set of gauged specimens that would be tested to validate the optical crack growth measurement method.

One other concern of the visual measurement of crack growth on a hand-drawn set of tick marks is that only one side of the specimen can be monitored. It is assumed that the crack front is perpendicular to the direction of crack growth and is relatively linear, but this has not yet been confirmed. A second observer on the back side of the DCB sample can verify that the crack front is symmetric, but this cannot confirm linearity. A series of partially-broken DCB samples may be C-scanned to determine the exact crack front shape in the specimen. Consistency in test-derived G_{Ic} values, which rely upon consistent crack growth measurement (Equation 1) within a specimen and from test to test suggest that the current method of crack tip measurement is adequate, though there are more elegant methods.

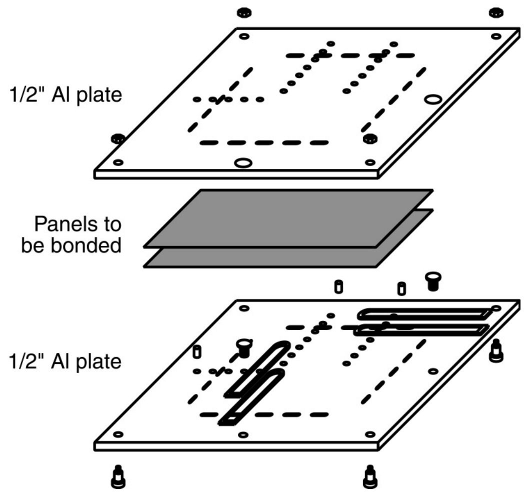


Figure 6. Exploded view of bonding jig.

Specimen Preparation: Bonding Jig

After evaluating the manual alignment method for the first two sets of bonded DCB specimens and the hinges applied to them, it was decided to create a bonding jig (Figure 6). The alignment of the bonded unidirectional laminates and the hinges that are held in the test machine grips can affect the strain energy release rates considerably. If bonded panels are positioned by hand and then held under weight, there is much opportunity for misalignment.

The bonding jig that was designed and constructed is essentially a large press made of two 18" x 18" x ½" aluminum 6061 plates. Features include:

1. 1/8" thick silicone sheets attached to the aluminum plates to distribute the press load evenly over local panel contours and prevent squeezed-out adhesive from bonding the entire jig together.
2. A grid of peg holes to allow panels to be aligned and centered in the jig.
3. Two pairs of staggered clamping sliders to keep the panels against the alignment pegs, even with samples that are uneven or unequal in size.
4. 20 pairs of slots to accommodate hinge pins for aligning hinges to DCB samples and for bonding both hinges to a specimen simultaneously.

Specimen Preparation: Bondline Thickness Control

The bond thickness for the first two sets of DCB samples was controlled by mixing in 2.5% by weight of 0.005" diameter glass microbeads into the epoxy before bonding. Because the two-part epoxy was mixed by hand and because the microbeads visually disappear once mixed in, it was impossible to determine if the beads were distributed evenly.

While examining fracture surfaces with a scanning electron microscope (SEM), it was found that there were fine separation layers between the glass microbeads and the adhesive (Figure 7). This indicates that the adhesive did not bond to the microbeads, creating possible crack nucleation points, potentially weakening the

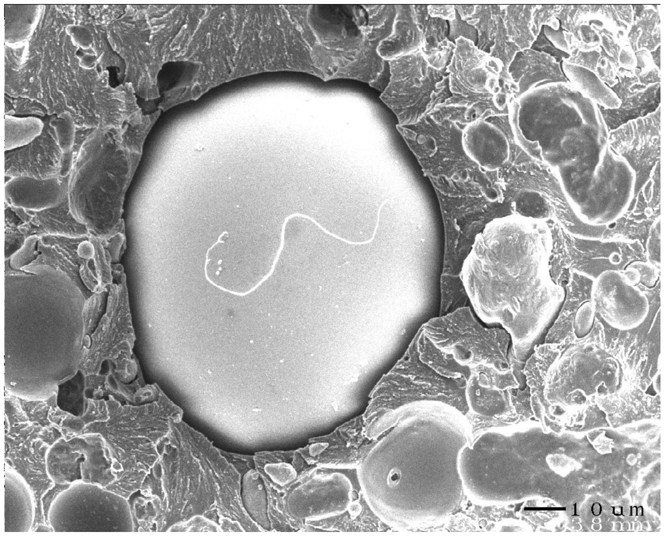


Figure 7. 0.005” diameter glass bead in adhesive on DCB fracture surface. 429× magnification.

overall bond and lowering the measured G_{Ic} values. However, alternate theories propose that this lack of bond between the glass beads and the adhesive may actually lead to crack blunting, thereby increasing G_{Ic}.

Pre-treatment of the glass beads with a silane adhesion promoter will ensure proper bonding between the beads and the adhesive, but it will not address the distribution issue. Alternate spacing methods are under consideration, including distributed wires, tabs, and layers of tape. These alternate methods allow more control over distribution, but care must be taken to compensate for these large embedded features when cutting bonded panels into individual specimens.

Additionally, the SEM photos revealed what appear to be very small cavities created by air trapped in the mixed adhesive, another potential crack initiation site. In the future, mixed two-part epoxy may be placed in a vacuum chamber prior to application on the panels to be bonded.

CONCLUSIONS

Because DCB and wedge test results accurately predict short-term strength and long-term adhesive bond durability in service, respectively, surface preparation methods that affect bond strength are evaluated. DCB testing suggests that grit blasting surfaces prior to bonding led to higher G_{Ic} values, though the mode of failure (interfacial or cohesive) is unchanged from a non-blasted sample. Adhesive bonding to composite surfaces that were cured against a nylon peel ply rather than a PTFE vacuum bag film showed the following trends:

1. Failure at lower loads and opening displacements.
2. Intermittent crack propagation.
3. Lower G_{Ic} values.
4. Interfacial, not cohesive failure.

Wedge tests, in an elevated temperature and humidity environment, with the same custom bonded specimen, will utilize DCB test results to predict joint failure.

Due to geometric constraints and the low in-plane fracture strain of composites materials, floating roller tests were found unsuitable for testing bonded composites for the thicknesses used herein. A bonding jig was designed and constructed to ensure accurate alignment and bonding of specimens and DCB hinges.

REFERENCES

1. Hart-Smith, L. J. 1996. "The Curse of the Nylon Peel Ply," presented at *41st International SAMPE Symposium*, March 24-28 1996, pp. 303-317.
2. Bascom, Willard D. and Cottington, Robert L. 1976. "Effect of Temperature on the Adhesive Fracture Behavior of an Elastomer-Epoxy Resin," *Journal of Adhesion*, 20: 333-346.
3. Marceau, J. A., Moji, Y., and McMillan, J. C. 1976. "A Wedge Test for Evaluating Adhesive Bonded Surface Durability," *21st National SAMPE Symposium*, Los Angeles, CA, April 6-8, 1976, pp. 332-355.
4. Cognard, J. 1987. "Quantitative Measurement of the Energy of Fracture of an Adhesive Joint Using the Wedge Test," *Journal of Adhesion*, 22(2): 97-108.
5. Cognard, J. 1986. "The Mechanics of the Wedge Test," *Journal of Adhesion*, 20(1): 1-13.
6. Crosley, P. B., and Ripling, E. J. 1991. "A Thick Adherend, Instrumented Double-Cantilever-Beam Specimen for Measuring Debonding of Adhesive Joints," *Journal of Testing and Evaluation*, 19 (1): 24-28.
7. Ripling, E. J., Mostovoy, S., Bersch, C. 1971. "Stress Corrosion Cracking of Adhesive Joints," *Journal of Adhesion*, 3: 145-163.
8. Sloan, Forrest. 1993. "A Constant-*G* Double Cantilever Beam Fracture Specimen for Environmental Testing," *Journal of Composite Materials*, 27(16): 1606-1615.
9. Johnson, W. S., and Butkus, L. M. 1988. "Considering Environmental Conditions in the Design of Bonded Structures: A Fracture Mechanics Approach," *Fatigue & Fracture of Engineering Materials & Structures*, 21(4): 465-478.
10. Hart-Smith, L. J. 1997. "A Peel-Type Durability Test Coupon to Assess Interfaces in Bonded, Co-Bonded, and Co-Cured Composite Structures," McDonnell Douglas Paper MDC 97K0042, presented to *MIL-HDBK-17 Meeting*, Tucson, April 14-17, 1997.
11. Jurf, R. A. 1988. "Environmental Effects on Fracture of Adhesively Bonded Joints," *Adhesively Bonded Joints: Testing, Analysis, and Design,* ASTM STP 981, pp. 276-288.
12. Chai, Herzl. 1986. "Bond Thickness Effect in Adhesive Joints and its Significance for Mode I Interlaminar Fracture of Composites," presented at *Composite Materials: Testing and Design (Seventh Conference)*, ASTM STP 893, J. M. Whitney, Ed., pp. 209-231.
13. Chang, D. J., Muki, R., and Westmann, A. 1976. "Double Cantilever Beam Models in Adhesive Mechanics," *International Journal of Solids and Structures*, 12(1): 13-26.
14. Mostovoy, S., Ripling, E. J., Bersch, C. F. 1971. "Fracture Toughness of Adhesive Joints," *Journal of Adhesion*, 3: 125-144.
15. Penado, F. E. 1993. "A Closed Form Solution for the Energy Release Rate of the Double Cantilever Beam Specimen with an Adhesive Layer," *Journal of Composite Materials*, 27(4): 383-407.
16. Ripling, E. J., Mostovoy, S., and Corten, H. T. 1971. "Fracture Mechanics: A Tool for Evaluating Structural Adhesives," *Journal of Adhesion*, 3: 107-123.
17. El-Senussi, A. K., and Webber, J. P. H. 1984. "On the Double Cantilever Beam Technique for Studying Crack Propagation," *Journal of Applied Physics*, 56(4): 885-889.
18. Whitney, J. M., Browning, C. E. 1982. "A Double Cantilever Beam Test for Characterizing Mode I Delamination of Composite Materials," *Journal of Reinforced Plastics and Composites*, 1: 297-313.