

The Evaluation of Aluminum Foil-Interlayered High-Modulus K13CU Pitch Fiber-Siloxane- Modified Resin Composite Panels

15 July 2003

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Prepared for

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20031126 093

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This report was submitted by The Aerospace Corporation, El Segundo, CA 90245-4691, under Contract No. F04701-00-C-0009 with the Space and Missile Systems Center, 2430 E. El Segundo Blvd., Los Angeles Air Force Base, CA 90245. It was reviewed and approved for The Aerospace Corporation by P. D. Fleischauer, Principal Director, Space Materials Laboratory. Michael Zambrana was the project officer for the Mission-Oriented Investigation and Experimentation (MOIE) program.

This report has been reviewed by the Public Affairs Office (PAS) and is releasable to the National Technical Information Service (NTIS). At NTIS, it will be available to the general public, including foreign nationals.

This technical report has been reviewed and is approved for publication. Publication of this report does not constitute Air Force approval of the report's findings or conclusions. It is published only for the exchange and stimulation of ideas.


Michael Zambrana
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REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 0704-0188

Public reporting burden for this collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing this collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggestions for reducing this burden to Department of Defense, Washington Headquarters Services, Directorate for Information Operations and Reports (0704-0188), 1215 Jefferson Davis Highway, Suite 1204, Arlington, VA 22202-4302. Respondents should be aware that notwithstanding any other provision of law, no person shall be subject to any penalty for failing to comply with a collection of information if it does not display a currently valid OMB control number. PLEASE DO NOT RETURN YOUR FORM TO THE ABOVE ADDRESS.

1. REPORT DATE (DD-MM-YYYY) 15-07-2003		2. REPORT TYPE		3. DATES COVERED (From - To)	
4. TITLE AND SUBTITLE The Evaluation of Aluminum Foil-Interlayered High-Modulus K13CU Pitch Fiber-Siloxane-Modified Resin Composite Panels				5a. CONTRACT NUMBER F04701-00-C-0009	
				5b. GRANT NUMBER	
				5c. PROGRAM ELEMENT NUMBER	
6. AUTHOR(S) R. J. Zaldivar and R. Casteneda				5d. PROJECT NUMBER	
				5e. TASK NUMBER	
				5f. WORK UNIT NUMBER	
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) The Aerospace Corporation Laboratory Operations El Segundo, CA 90245-4691				8. PERFORMING ORGANIZATION REPORT NUMBER TR-2003(8565)-5	
9. SPONSORING / MONITORING AGENCY NAME(S) AND ADDRESS(ES) Space and Missile Systems Center Air Force Space Command 2450 E. El Segundo Blvd. Los Angeles Air Force Base, CA 90245				10. SPONSOR/MONITOR'S ACRONYM(S) SMC	
				11. SPONSOR/MONITOR'S REPORT NUMBER(S) SMC-TR-04-03	
12. DISTRIBUTION/AVAILABILITY STATEMENT Approved for public release; distribution unlimited.					
13. SUPPLEMENTARY NOTES					
14. ABSTRACT A contractor is currently fabricating flat panels of high-modulus fiber K13CU/siloxane-modified polycyanurate matrix resin composites. Mechanical testing at a commercial lab showed that the interlaminar shear strengths and the flat-wise tensile properties were below that expected. We evaluated the degree of consolidation and cure to verify that incorporated foil layers were not causing premature failure. A number of composite panels were analyzed using dynamic mechanical analysis and optical microscopy. The measured glass-transition temperatures (Tg's) of the samples all suggested that they were properly cured. A solvent exposure test developed in our laboratory also corroborated these results. Optical micrographs of cross-sections for the different composites indicated good consolidation and no observable foil debonds. All of the samples exhibited similar failure modes from our testing. The focus of the lower-than-expected strengths was then placed on the sample preparation procedure for mechanical testing. Sample preparation of the test specimens at the testing laboratory was identified as the cause for the lower-than-expected strengths. Subsequent tests on articles with proper sample preparation yielded acceptable mechanical strengths.					
15. SUBJECT TERMS Composite, Siloxane, Fibers, K13Cu, Glass transition temperature, Cyanate ester, Piolycyanurate.					
16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT	18. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON
a. REPORT	b. ABSTRACT	c. THIS PAGE			Rafael Zaldivar
UNCLASSIFIED	UNCLASSIFIED	UNCLASSIFIED		11	19b. TELEPHONE NUMBER (include area code) (310)336-1999

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1. Introduction

The siloxane-modified polycyanurate resin system has been developed for use as the matrix material for satellite composite structures. This resin system has a very low moisture absorption behavior, which contributes to excellent dimensional stability of the structure. However, additional measures are necessary to prevent moisture uptake due to its sensitivity during processing. A fabricator has developed and manufactured a series of high-modulus fiber K13CU composite panels that are cured with aluminum foils interlayed within the composite. An aluminum foil is placed one ply from the outer surface, on both sides. Once all of the prepregs have been stacked, the composite is then bagged and cured (see Figure 1). The resin within the prepreg forms an adhesive bond to the foil material. The aluminum layer reduces moisture uptake by preventing diffusion from the outer surfaces into the composite. This, in turn, reduces the coefficient of moisture expansion for the part. Preliminary mechanical tests at a commercial mechanical testing laboratory have indicated that the interlaminar shear strength and the transverse tensile are below what is expected for this fiber-matrix combination. Previous investigations on other configurations with the foil material have indicated no failure at the aluminum-resin interface. This indicates that the nominal bond should not be degraded by the introduction of the foil.

As discussed in previous reports,^{1,2} the cyanate ester base resin systems are extremely susceptible to moisture contamination and hydrolysis during cure. Even low concentrations of moisture during cure have been shown to sharply reduce the glass-transition temperature (T_g) of the resin as well as degrade their mechanical performance. The objective of this report is to determine whether the degree of cure and/or poor consolidation of these composites could have affected the mechanical properties.

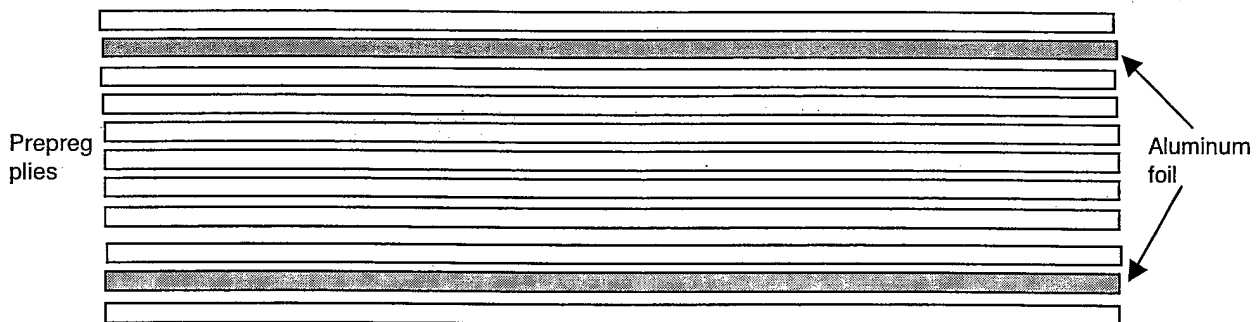


Figure 1. Schematic showing aluminum-foil layers interlayed within the ply lay-up.

2. Experimental

2.1 Materials

Four composite panels were obtained from the fabricator. All of the specimens were 2.0 in. x 2.0 in. Three of the specimens appeared to be the same thickness with an aluminum foil interlayer one ply from the outer surface. The fourth specimen was 0.25 in. in thickness and appeared to have 4 layers of interlayered aluminum foil within the composite. Two of the foils were one ply from the outer surface and the other two plies were in the center of the composite, balanced on either side of the center ply.

2.2 Dynamic Mechanical Analysis

A Rheometrics Dynamic Mechanical Analyzer (DMA) was used to determine the T_g of all composite samples. The DMA subjects a sample to cyclic torsional deformations and quantifies the material response by measuring the shear modulus, G' , the shear loss, G'' , and the lag angle between the applied stress and resulting strain, $\tan \delta$, as a function of temperature. In this report, the peak of the G'' curve was used for determination of the T_g values.

2.3 Optical Microscopy

Transverse polished cross sections of the tested samples were prepared by mounting the sample in a room-temperature-cured epoxy and grinding and polishing to a 1- μm finish with diamond paste. The cross sections were examined by optical microscopy with a Nikon Epiphot metallographic microscope equipped with a SONY DXC-107A CCD video camera.

2.4 Solvent exposure

Cubes 0.5 x 0.5 x 0.25 in. were submerged in equal amounts of tetrahydrofuran solvent and allowed to sit for one hour. The samples were then removed and checked for any debonding or ply separation.

2.5 Tensile Tests

Composite cross-sections 1.0 in. x 1.0 in. were bonded to 0.75-in. pinned aluminum rods. The composites were lightly sanded with 250-grit paper on their surfaces. The aluminum substrate was grit blasted and wiped with solvent prior to bonding. Dow 7640 epoxy adhesive was used for all bonding procedures. The samples were loaded in a Universal Instron at a rate of 0.05 in./min. A 5000-lb load cell was used for all testing.

3. Results

Numerous flat laminates of K13CU/siloxane-modified polycyanurate composites have been investigated in our laboratory. The K13CU fiber is a pitch-based carbon fiber that is very difficult to work with, especially during the prepregging procedure, due to its exceedingly high modulus. In addition, the laminates fabricated for this application also incorporated aluminum interlayer foils to resist moisture diffusion, further complicating the process. The same aluminum layers that are incorporated into the composite to prevent moisture diffusion inward complicate the removal of volatiles during consolidation. Table 1 shows Tg values for some of the initial laminates fabricated. As shown, the majority of the composites appear to be well cured. Typically, a glass-transition temperature above 170°C for this resin system produces a fairly good product. However, a Tg value a few degrees below 170°C, indicates insufficient cure that strongly degrades the resin's mechanical properties by producing a brittle, low strain-to-failure matrix material.

Optical microscopy of the initial lots of composite parts shown in Table 1 indicated that a high degree of interlayer porosity was located throughout the samples. Figures 2 and 3 show cross-sections of a typical K13CU laminate. The black areas in the micrograph represent areas of voids. The lighter areas of gray and white denote the matrix and fiber. As shown, there are substantial void areas oriented along the ply orientation. This is typical of a sample with poor resin flow characteristics and trapped volatiles. However, in this case, it may also be due to the nature of the materials used and the aluminum foil interlayer barrier as shown in Figure 3. As might be expected, the foil may behave as a barrier to the volatiles during cure. The mechanical properties of this composite would be severely degraded, especially in the matrix-dominated directions. A number of composite manufacturing trials were performed by the contractor to eliminate or reduce the porosity and improve consolidation. The set of samples shown in Table 2 were manufactured once the contractor optimized all of the processing issues.

Table 2 shows the four different composites investigated in this study. All of the composites were tested using dynamic mechanical analysis, and the glass temperatures are shown. As shown, the Tg's (measured from the peak of the loss modulus curve) for all of the samples were within family and indicate full cure. No indication of contamination was observed in any of the samples. The samples

Table 1. Glass-Transition Temperature (Tg) Values for K13CU Laminates

Lot number	Tg (°C)	Solvent Exposure
(1) H047PB001-001 SN 010	171	Pass
(2) H047PB001-001 SN 009	171	Pass
(3) H047PB003-002 SN 001	171	Pass
(4) H047E003-001 SN 003	170	Pass
(5) H047PB003-002 SN-002	171	Pass
(6) H047PB003-002 SN-003	170	Pass
(7) H047E003-001 SN 002	168	Fail

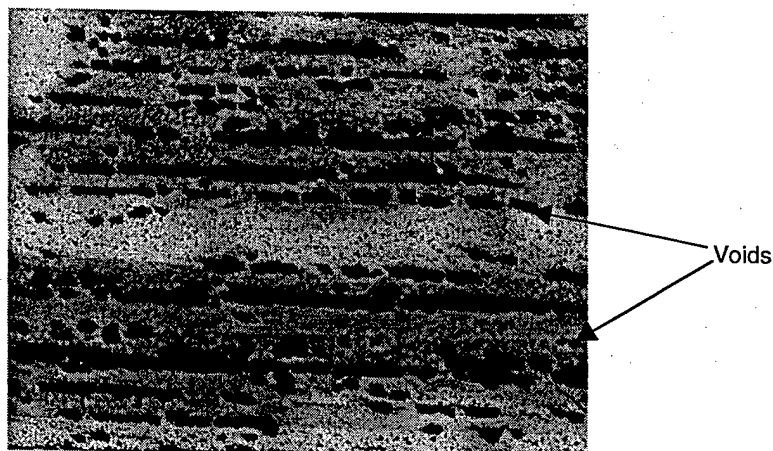


Figure 2. Optical Micrograph showing midplane cross-section of K13CU laminate. Micrograph shows a large degree of interply porosity located between plies.

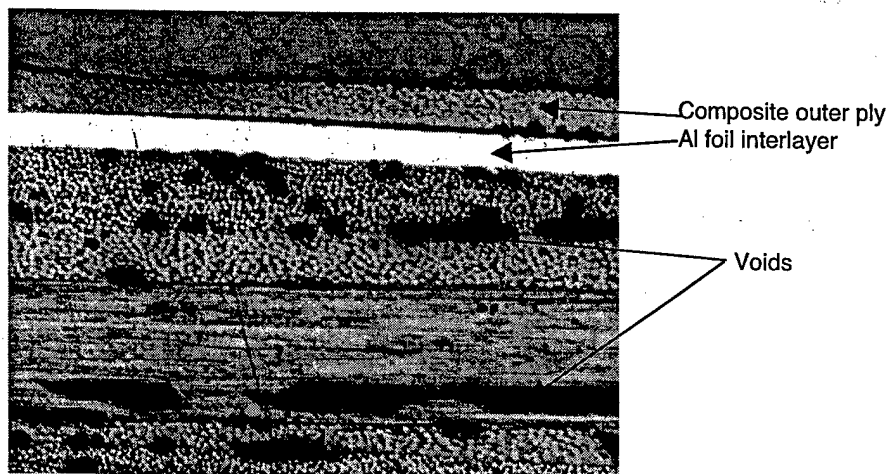


Figure 3. Optical Micrograph showing higher magnification cross-section of K13CU laminate with aluminum-foil interlayer at outer surface.

Table 2. Tg Values for Several Composites Investigated in This Study.

Composite Sample I.D.	Tg (DMA, G ^o)	Solvent Exposure
Polycarbonate standard	152°C	-
H047PB004-001-004 Sample (1)	175°C	No degradation
H047E002-002 S/N 008 Sample (2)	173°C	No degradation
H047PB004-001-003 Sample (3)	174°C	No degradation
H047PB004-005 S/N 001 Sample (4)	173°C	No degradation

were also exposed to THF solvent for 1 h. Typically, if the entire system or selected areas of the composite are partially cured, solvent exposure will cause degradation of the part. A well-cured system will resist degradation. In a previous report, THF solvent exposure was shown to identify par-

tially cured or contaminated systems. None of the samples investigated in this study exhibited degradation.

Figures 4-9 show cross-sections of the composites investigated in this study. Figures 4a and b are optical micrographs of sample 1. The white areas are the carbon fibers and the gray area represents the resin matrix material. The variations shown in the fiber cross-sectional area are due to changes in orientation of the plies. As shown, the composite appears well consolidated throughout. However, the small black specks distributed randomly throughout are indicative of closed porosity in the structure. The fiber volume of the outer ply appears to be in the 50% range that is lower than the 60% fiber volume range of the overall composite. However, the purpose of the outerply is more cosmetic than structural.

The aluminum-foil layer can be seen one ply in from the outer surface. Figure 4a shows this layer. The foil appears well bonded to both sides of neighboring laminate plies. Figure 5a shows a higher magnification photo of the aluminum foil within the structure of sample 1. No areas of debonding or excessive fiber-to-metal contact were

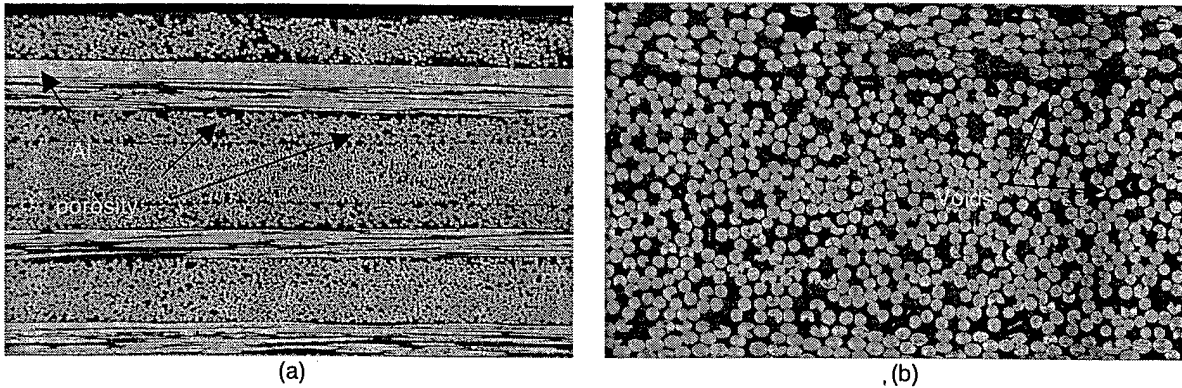


Figure 4. Cross-sectional optical micrograph of composite sample 1. The composite appears fairly well consolidated with some porosity observed within plies.

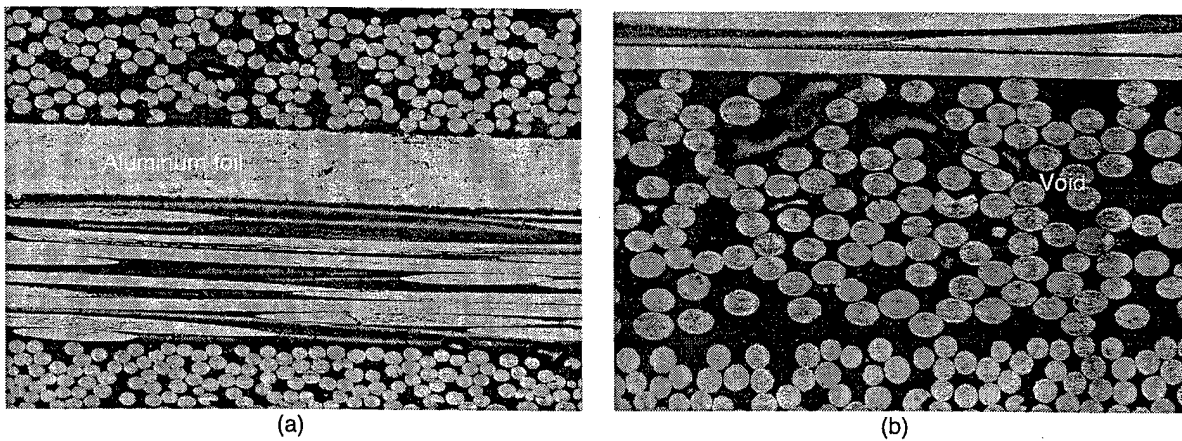


Figure 5. Cross-sectional optical micrograph of composite sample 1. (a) Shows good bonding on aluminum foil in the structure; (b) shows porosity within plies.

observed. Resin-lean areas would account for decreases in interlaminar shear strength; however, the sample appeared fairly resin rich on both sides. Figure 5b shows a higher magnification micrograph of the porosity observed within plies. The porosity appeared fairly well distributed throughout. Image analysis indicated a void volume of less than 2%.

Figures 6 and 7 show cross-sections of sample 2. As shown, there appears to be consolidation and minimal porosity within the plies of this composite also. The aluminum-foil interface appears to be well bonded to the adjacent plies by the matrix material. No indications that would result in decreased mechanical properties were identified.

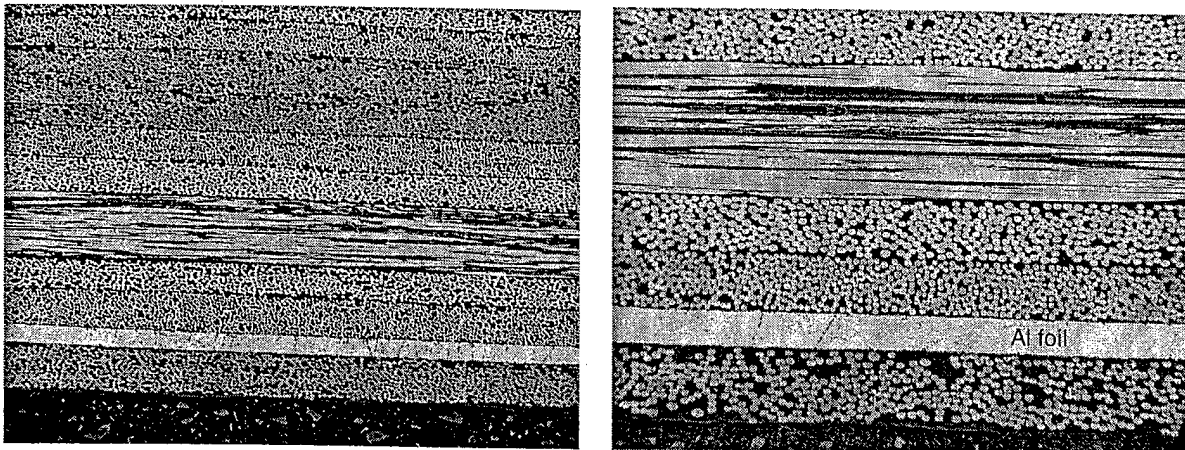


Figure 6. Cross-sectional optical micrograph of composite sample 2. (a) Shows good consolidation to aluminum foil in the structure; (b) shows low fiber content on outer ply.

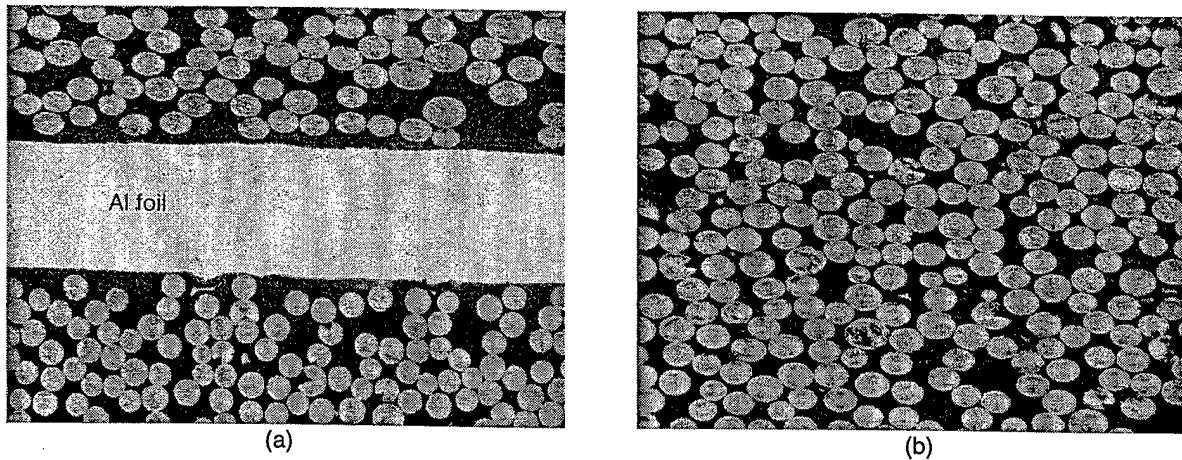


Figure 7. Cross-sectional optical micrograph of composite sample 2. (a) shows good bonding on aluminum foil in structure; (b) shows minimal porosity within plies.

Optical micrographs of microstructural cross-sections of samples 3 and 4 are shown in Figures 8 and 9. Composite sample 3 appeared to have some pronounced porosity between the second and third ply. However, the porosity was limited to a fraction of the entire length of the sample. There were no indications of debonding between the foil layer and the adjacent prepreg. Composite sample 4 was notably different from the other specimens. Two aluminum layers were positioned in the mid-plane of the sample, in addition to the two foil layers closer to the sample surface. Sample 4 exhibited no other differences, which would raise cause for concern. Mechanical tests were then performed on the tag specimens to evaluate the failure mode.

Composite samples were tested in tension perpendicular to the ply direction. The samples' surfaces were roughened and bonded to aluminum plugs as described in the experimental section. It is critical to achieve a strong bond between the adhesive and the composite material to evaluate composite

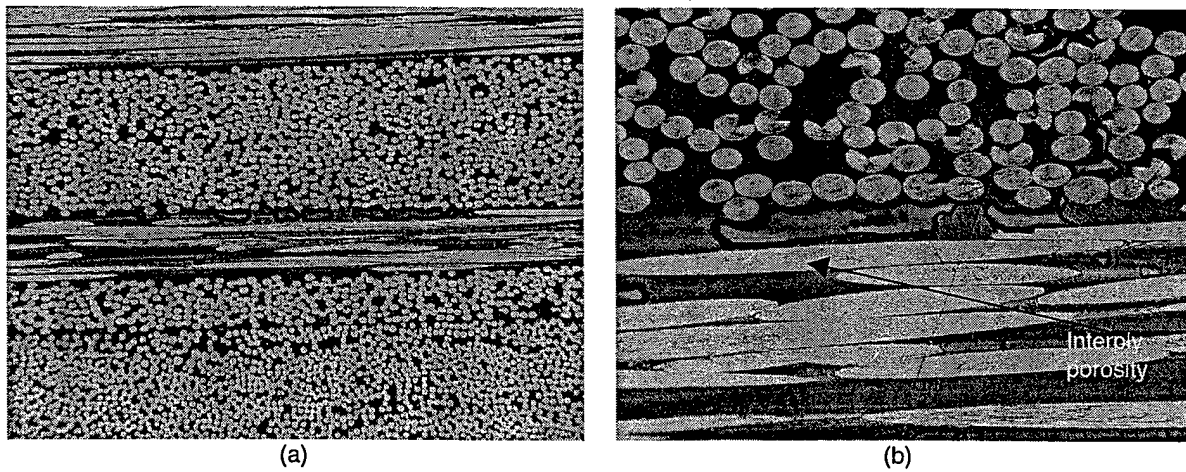


Figure 8. Cross-sectional optical micrograph of composite sample 3. (a) shows good bonding on aluminum foil in the structure; (b) shows porosity within plies.

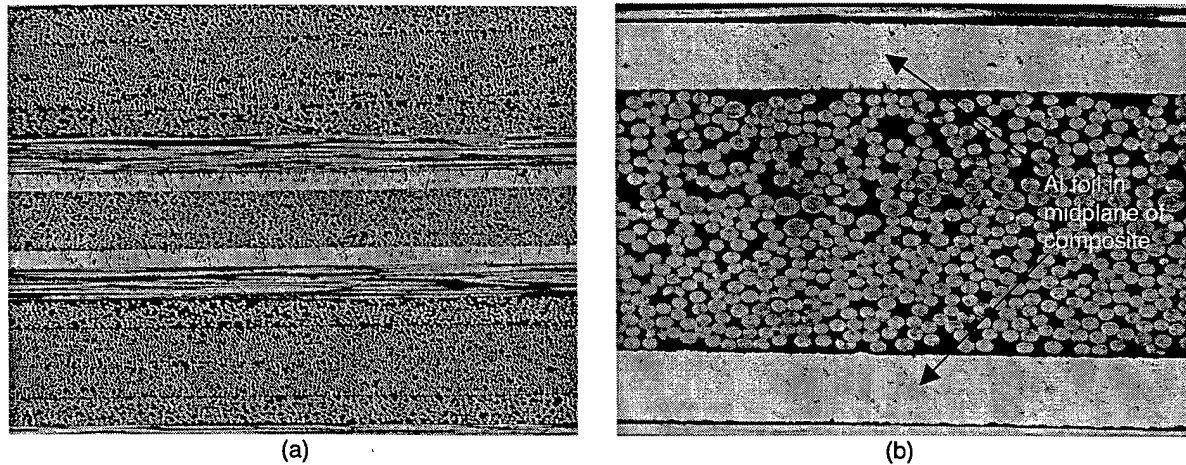


Figure 9. Micrograph of composite sample 4. (a) shows good bonding on aluminum foil in the structure; (b) shows minimal porosity within plies.

interply performance. The main reason for this testing was not to acquire mechanical strengths, but rather to determine where the failure was occurring. Figure 10 shows the fracture face for a typical flatwise tension specimen. This sample shows a mixed mode. Over 60% of the sample has failed within the composite, exhibiting a ply-to-ply separation. The majority of the failures occur within the first 3 outer plies. In this case, there appears to be some exposed metal from the aluminum plug. This indicates that some of the failure occurred at the interface between the aluminum plug and the bonding epoxy. An ideal sample failure would exhibit no such separation or adhesive failure. The failure load for this sample was somewhat decreased due to this separation. The other 3 specimens exhibited complete ply-to-ply separation.

All of the materials and processing issues appeared to be fine; therefore, the focus of the lower-than-expected strengths was placed on the test laboratory subcontractors specimen preparation procedure. The contractor was notified of our results, and we asked about the failure mode being experienced by the majority of the samples. Only one of our samples out of all of the lots exhibited an aluminum foil to composite separation. The subcontractor analyzed the failed samples and observed that the majority of the failures were actually occurring on the outer surface between the bonding adhesive and the composite face. This failure mode is usually indicative of poor sample preparation. Lower-than-expected strengths would be expected from this type of behavior. In order to improve the bond and get the correct type of failure mode, the composite sample needs to be roughened considerably to ensure a strong mechanical bond with the bonding adhesive. In addition, the type of high-modulus, pitch-based carbon fiber used in this composite is prone to weaker bonding than lower modulus PAN-based fiber typically used. The contractor instructed the testing laboratory in correct sample preparation, and, thus far, all of the composite test specimens appear to be failing within allowable ranges and within the composite.



Figure 10. Fracture surface of sample 1, 2, 3, and 4 all appeared to fail within the composite.

4. Conclusion

- The glass-transition temperatures of all four composites were between 172–175°C, indicating that full cure was achieved during processing.
- Optical microscopy of all of the composite samples indicated that good consolidation of the plies was achieved. Fiber volumes also appeared consistent, and the aluminum-foil interlayer did not appear to prevent volatile evolution. Some porosity was observed throughout the samples; however, it appeared to be within the acceptable limits (<2%) and was not segregated to any one region.
- The aluminum-foil interlayers within the composite appeared to be well bonded and consolidated within the structure. No areas of debonding or excessive fiber-to-metal contact were observed.
- Flatwise tensile samples of our composites were performed to verify the failure mode. Three of the four samples exhibited composite ply-to-ply failure as expected. One of the samples (H047PB004-005 S/N 001) exhibited a complete aluminum-foil interfacial failure. Additional testing of this sample is necessary to verify whether there is a need for concern.
- There appeared to be no issues dealing with the composite material investigated that would cause degradation in flatwise tension strengths. We suggest that the contractor investigate the sample preparation process that the laboratory testing subcontractor is using as well as verify the failure mode of all tested samples.

LABORATORY OPERATIONS

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