ADHESIVE BONDING CHARACTERIZATION OF COMPOSITE JOINTS FOR CRYOGENIC USAGE

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ABSTRACT

The development of polymer composite cryogenic tanks is a critical step in creating the next generation of launch vehicles. Future reusable launch vehicles need to minimize the gross liftoff weight (GLOW). This weight reduction is possible due to the large reduction in weight that composite materials can provide over current aluminum technology. In addition to composite technology, adhesively bonded joints potentially have several benefits over mechanically fastened joints, such as weight savings and cryogenic fluid containment. Adhesively bonded joints may be used in several areas of these cryogenic tanks, such as in lobe-to-lobe joints (in a multi-lobe concept), skirt-to-tank joint, strut-to-tank joint, and for attaching stringers and ring frames. The bonds, and the tanks themselves, must be able to withstand liquid cryogenic fuel temperatures that they contain. However, the use of adhesively bonded composite joints at liquid oxygen and hydrogen temperatures is largely unknown and must be characterized. Lockheed Martin Space Systems Company, Michoud Operations performed coupon-level tests to determine effects of material selection, cure process parameters, substrate surface preparation, and other factors on the strength of these composite joints at cryogenic temperatures. This led to the selection of a material and process that would be suitable for a cryogenic tank.

KEY WORDS: Composites, Adhesive Bonding, Cryogenics

1. INTRODUCTION

Lockheed Martin Space Systems Company, Michoud Operations (hereafter known as LM Michoud) has been a leader in cryogenic tank technology. LM Michoud has been fabricating external tanks for the Space Shuttle for over 25 years. For X-33, Lockheed Martin VentureStar, X-34, and other future launch vehicles, LM Michoud is applying this expertise towards the development of lightweight composite cryogenic tankage.

These next-generation launch vehicles must reduce dry lift-off weight in order to increase payload capability and reduce payload cost per pound. One way to meet this goal is to fabricate composite tanks, which can potentially reduce weight up to 40% in certain vehicle designs. Adhesive bonding of these tanks is also a weight-savings opportunity. In addition, adhesively bonded joints can eliminate leak paths associated with through fasteners.

The issue with adhesively bonded composite joints is that they are unproven at cryogenic temperatures, such as those associated with liquid hydrogen and liquid oxygen. Also, the tanks in many vehicle designs, while at –252°C (-423°F) when filled with liquid hydrogen, may see temperatures of +121°C (+250°F) or higher during re-entry. The adhesive selected for use must be able to maintain properties throughout this temperature range. LM Michoud implemented a test plan at the coupon level to characterize adhesives for cryogenic use.

2. EXPERIMENTAL

LM Michoud laid out the test plan in several phases. Phase 1: Preliminary Testing was used to identify factors affecting performance such as specimen geometry and surface preparation. Phase 2: Adhesive Screening was designed to compare several different adhesives. Phase 3: Bond Design Characterization looked at such bond design factors as overlap length and near ply orientation on adhesive bond strength.

2.1 Phase 1: Preliminary Testing: The goal of Phase 1 testing was to determine the optimal coupon preparation techniques and configurations for maximum bond performance. For Phase 1, all specimen groups consisted of 2 sets of five coupons each. One set was tested at room temperature, the other at liquid helium temperature –252°C (-423°F). All specimen groups used the same composite material (IM7/977-2 unitape), epoxy film adhesive (AF191, 080K), cure cycle, and 16 ply quasi-isotropic substrate lay-up. Bond line thickness was controlled via the reinforcement in the film.

A 3.81 cm (1.5 inch) overlap length was chosen as a standard overlap length for this test matrix. A double-lap as opposed to single-lap was also chosen as a standard. However, two specimen configurations were studied. The first, called the "standard" configuration, was designed to compare with various double lap configurations in literature. The second configuration was designed to emulate the ASTM standard for metal joints. See Figures 1 and 2 for specimen configurations.

Figure 1: "Standard" Configuration

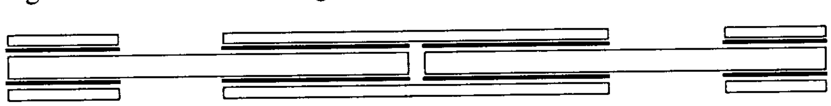

Figure 2: ASTM Configuration

Figure 3 shows the results of identically prepared specimens. The ASTM configuration had higher average strength values than the standard configuration, so the ASTM configuration was chosen as the configuration for future phases.

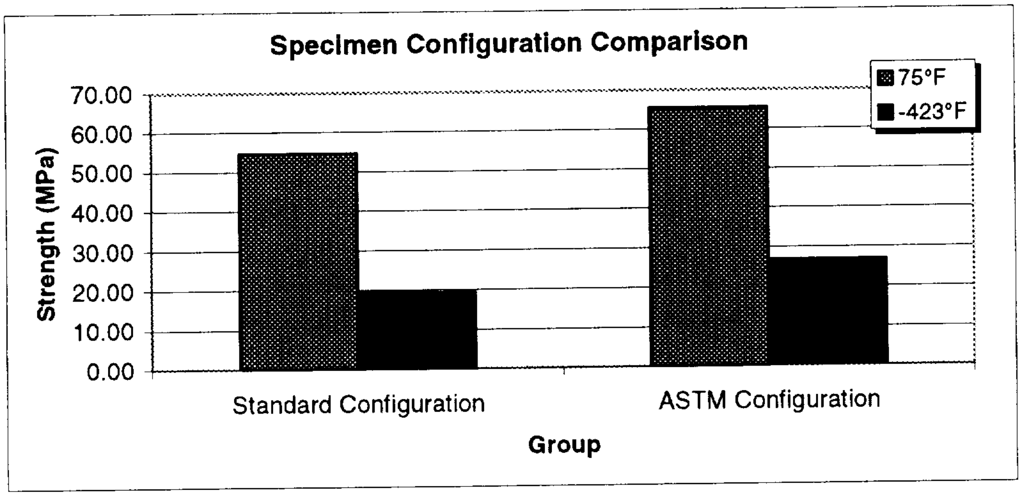

Figure 3: Comparison of Average Strength for the Standard and ASTM Specimen Configurations at Room Temperature and at Cryogenic Temperatures

Phase 1 also evaluated coupon preparation techniques. The two types of peel plies used were Super A, a nylon peel ply, and Super F, a polyester peel ply. For the "sanded" surfaces, surfaces were abraded with 150-180 grit aluminum oxide sandpaper using a multidirectional or circular sanding pattern. It was then sanded using 200-320 grit sandpaper in a multidirectional pattern. The samples were then wiped with isopropyl alcohol and wiped dry to remove dust and other contaminants. Silicon carbide sandpaper, Scotch-Brite, and/or other abrading materials were not used in this study. Table 1 identifies the groups and how they were prepared. Figure 4 shows the results of this phase of the study.

Table 1: Surface Preparation

Group	Description
AP1	No Peel Ply, Sanded
AP2	Super A Peel Ply, Sanded
AP3	Super A Peel Ply, Unsanded
AP4	Super A Peel Ply, Sanded, Primed
AP10	Super F Peel Ply, Sanded

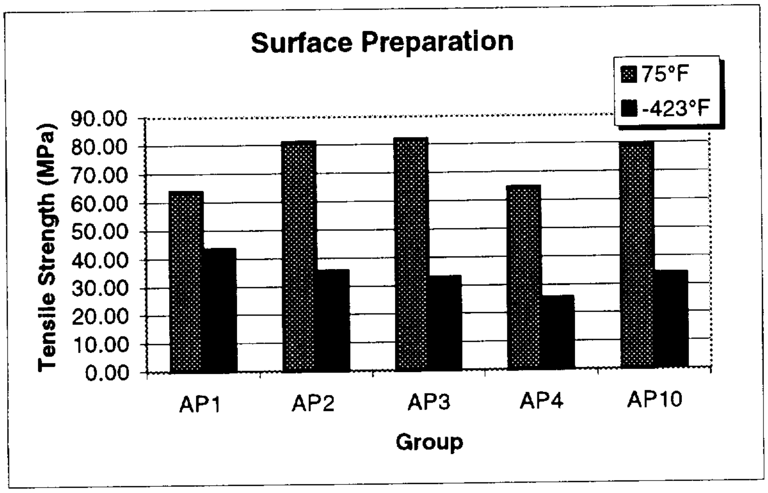

Figure 4: Surface Preparation Results

The results indicate little difference between the peel plies. The sample set that used an adhesive primer, typical for metals, on the composite substrate showed overall poor results. The effects of peel ply and sanding were different at the two temperatures. At cryogenic temperatures, substrates with no peel ply but that were sanded showed superior results. However, the best results at room temperature were on those samples that had a peel ply surface and that were not sanded. This difference in performance at the different temperatures was not expected and cannot be easily explained. For future phases, the compromise of Super A peel ply, sanded was selected as the baseline.

One phenomenon that was common between all Phase 1 tests was that for this particular adhesive, there was a severe drop in tensile strength between room temperature and cryogenic temperature. The cryogenic results showed a decrease between 32% and 65%, depending on the specimen group. This indicates that the cryogenic condition may be a critical design case for adhesive bonds subjected to these temperatures.

2.2 Phase 2: Adhesive Screening: The objective of the second phase of testing was to compare the performance of various adhesive types. Various adhesive chemistries and forms were selected for testing from a variety of manufacturers. All adhesive sets were cured per the manufacturer's recommendation. Once again, the same substrate material (IM7/977-2 unitape) and lay-up (16 ply quasi-isotropic) was used. Bond line thicknesses for the films were controlled via the reinforcement; while 10 mil beads were inserted into the paste adhesives to control thicknesses. All samples were the ASTM configuration, used Super A peel ply, and were sanded as described above. Table 2 provides a list of adhesives selected, while Figure 5 shows a comparison of performance.

Table 2: Screening Adhesives

Group	Name	Type
AS4	XEA 9361	Epoxy Paste
I	EA9394	Aluminum Filled Epoxy Paste
AS6	Crest 7450	Polyurethane Paste
AS7	1502-1	Cyanate Ester Paste
AS9	RS-4 Supported	Cyanate Ester Film
AS10	M2555	Cyanate Ester Film
AS12	FM 300-2 K(080)	Epoxy Film
AS13	1146 Film	Epoxy Film
AS14	Courtalds PR1665	Polyurethane Paste
AS17	HT424 Film	Aluminum Filled Epoxy/Phenolic

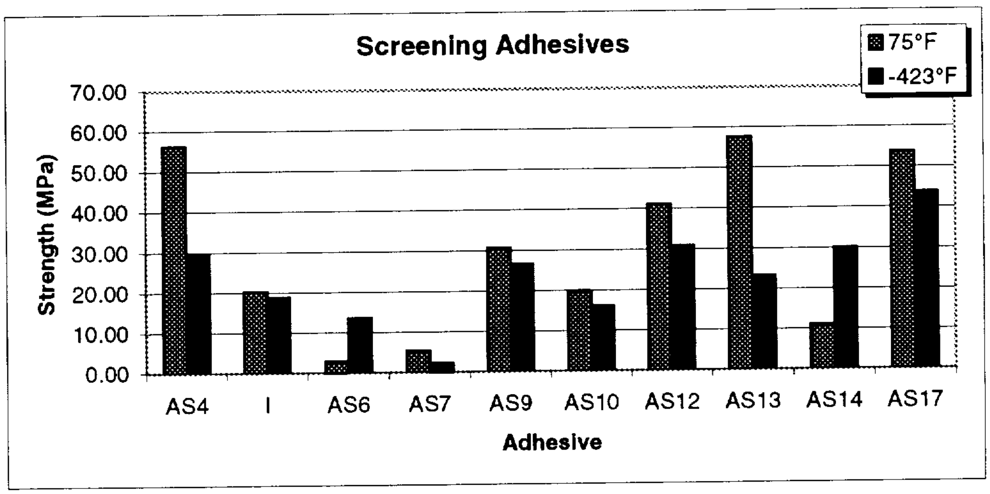

Figure 5: Screening Results

The first thing that stands out is that the drop in properties at cryogenic temperatures was not unique to the original baseline adhesive. Once again, the vast majority of adhesives showed a significant decrease in strength as a result of cryogenic temperature exposure. Only the two polyurethane paste adhesives showed an increase in properties at cryogenic temperatures. However, in both cases the room temperature results were uncharacteristically low, and elevated temperature properties are presumed to be even lower. Therefore, the polyurethane adhesives may not be acceptable for most launch vehicle tank joints.

The AS17 epoxy/phenolic adhesive did stand out; it had one of the highest room temperature values, and had very high cryogenic temperature performance. Also, the decrease between room temperature and cryogenic temperature was relatively meager at 18%.

In general, the film adhesives outperformed the paste adhesives, which was not unexpected. This is attributed to the presence of a support scrim in the adhesive, which likely helped control the bond line thickness and prevent excessive squeeze-out. It is also interesting to note that the cyanate ester adhesives performed poorly on the epoxy substrates. This was true regardless of form or manufacturer.

2.3 Phase 3: Bond Design Characterization: The objective of the third phase of testing was to determine the effects of various bond design parameters on the adhesive strength. These considerations included bond overlap length and near ply orientation.

For near ply orientation, the standard composite substrate was used. The only difference was that the plies, although still quasi-isotropic, had different ply orientations at the bond, either 45° or 90° (the previous specimen groups all had 0° at the near ply). For this study, three adhesives were selected: AF-191 080K (Adhesive 1), EA9394 with 10 mil beads (Adhesive 2), and HT424 Film (Adhesive 3). All were cured per the manufacturer's recommended cure cycle. The results are shown in Figure 6.

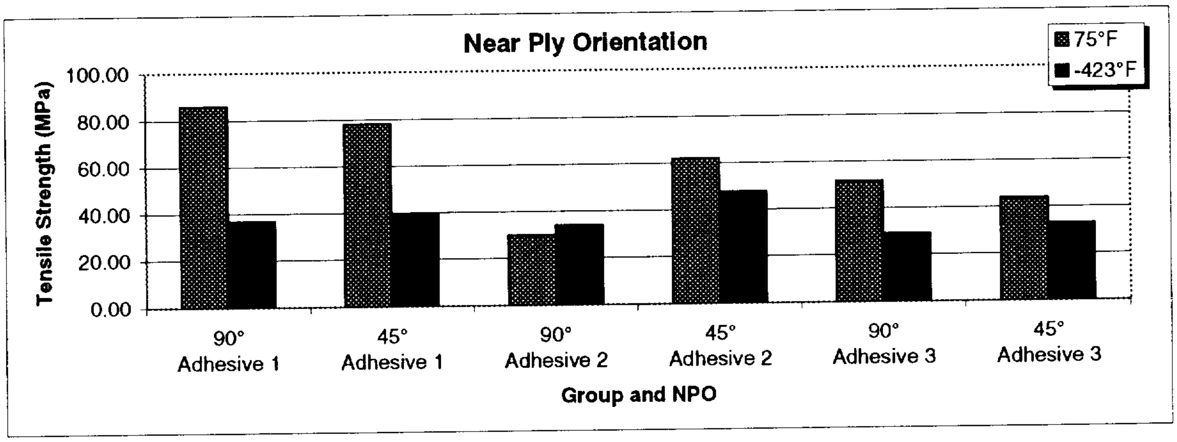

Figure 6: Near Ply Orientation

The effect of near ply orientation is minimal. A 45° ply at the bond line appears to be slightly better at cryogenic temperatures than a 90° ply. Nevertheless, the 90° ply appears superior for two out of the three adhesives at room temperature, all are within scatter, and are very close to the 0° value (Groups B, I, and AS-17, respectively, in previous figures). In conclusion, the near ply orientation does not appear to have a clear effect on bond performance.

Another design consideration was the effect of overlap length. In this study, the lap length was tested at various lengths up to 15.24 cm (6 inches). AF191-080K adhesive was used, and cured per the manufacturer's recommendation. The coupons were also tested at cryogenic (-252°C /-423°F), room temperature (75°F), and hot wet conditions. Hot wet samples were conditioned at 90% humidity for a minimum of one week and tested at 121° C (250°F). For each of the three conditions, the amount of load that the joint could handle increased with lap length, but the actual shear stress it could handle decreased with lap length. See Figures 7 and 8. However, at cryogenic and hot wet, there appeared to be "diminishing returns" in load capability after 7.62 cm (3 inch) lap length. Therefore, the 7.62-cm (3-inch) lap length appears to be the optimal balance for load capability and shear strength.

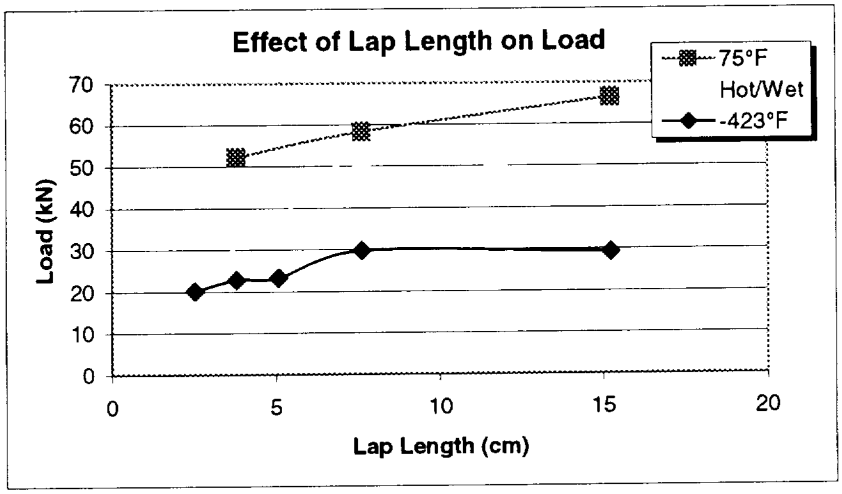

Figure 7: Lap Length vs. Load

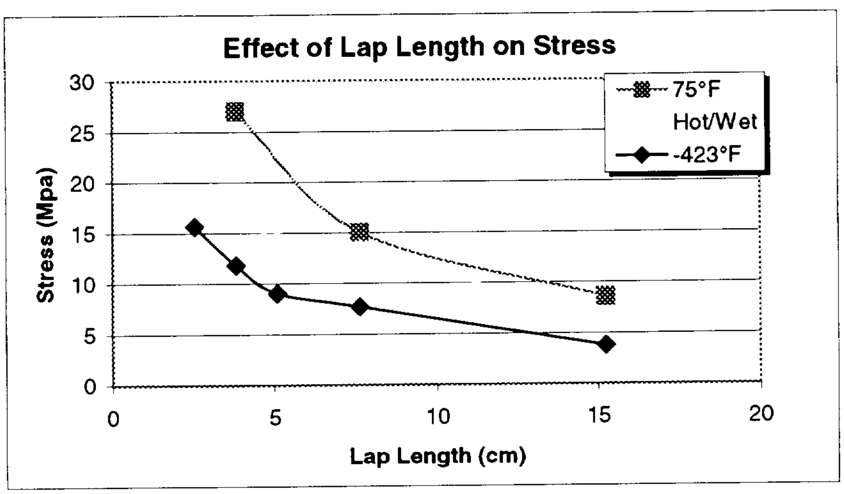

Figure 8: Lap Length vs. Stress

3. CONCLUSIONS

Lockheed Martin Michoud performed a comprehensive evaluation of adhesively bonded composite joints at cryogenic temperatures. The most significant result of the study is that for all adhesives tested (except polyurethanes), there was a significant decrease in lap shear strength at cryogenic temperatures. This held true regardless of specimen configuration, overlap length, surface preparation, etc. Therefore, when designing composite parts at cryogenic conditions with adhesive bonds, the cryogenic temperature must be considered a critical design parameter.