P66
EVALUATION OF CRYOGENIC PERFORMANCE OF ADHESIVES USING COMPOSITE-ALUMINUM DOUBLE LAP JOINTS

Sang-Guk KANG, Myung-Gon KIM, Chun-Gon KIM
Division of Aerospace Engineering, Department of Mechanical Engineering
Korea Advanced Institute of Science and Technology
373-1, Kuseong-dong, Yuseong-gu, Daejeon, 305-701, South Korea

ABSTRACT

In the development of a cryogenic propellant tank, the proper selection of adhesives to bond composite and liner is important for the safety of operation. In this study, 3 types of adhesives were tested for the ability to bond CFRP composites developed for cryogenic use and aluminum alloy (Al 6061-T6) for lining the tank using double-lap joint specimens. The double-lap joint specimens were tested inside an environmental chamber at room temperature and cryogenic temperature (-150°C) respectively to compare the bond strength of each adhesive and fracture characteristics. The material properties with temperature of component materials of double-lap joints were measured. In addition, ABAQUS was used for the purpose of analyzing the experimental results.

KEY WORDS: Joint Design/Joint Analysis, Joining/Joints/Bonding, Environmental Effects

1. INTRODUCTION

As the importance of space technology is emphasized, each nation is now actively doing research on futuristic technology for launch vehicles. Especially, the lightweight of the propellant tank that occupies much of the weight of launch vehicle has interested many researchers. For this reason, research has focused on substituting composite materials, which are excellent in specific stiffness and specific strength, for conventional tank materials [1-3]. Composites, however, can easily have micro cracks in matrix due to the difference in coefficients of thermal expansion

(CTE) of reinforcing fibers and matrix. These matrix cracks can be eventually related to phenomena of fuel leakage that is very dangerous for tank safety [4-5]. A metal liner is used with composites to make the type III tank since there are still many difficulties in using composites without a metal liner like type IV tank [6].

In the fabrication process of the metal-lined composite propellant tank, film or paste adhesive is applied between the composite and a metal liner. When the tank stores pressurized cryogenic propellant after the composite cryogenic tank is cured, the adhesive layer between the composite and a metal liner undergoes extreme thermal stress and may cause separation of these two materials. This imperils the safety of the tank seriously. Therefore, the research on selecting proper adhesives is required for manufacturing composite propellant tanks.

In general, tension test of a single-lap joint or a double-lap joint is often used to evaluate the bond strength of adhesives. Jianmei et al. [7], through the series of tensile tests of double-lap joints composed of some kinds of adhesives, evaluated the bond strength at low temperature and conducted finite element analysis to evaluate the effect of bond thickness and bending stress due to the eccentricity of a test setup on joint strength. Tong [8] modified the conventional failure prediction equation such as Tsai or Norris criterion and predicted the bond strength of a joint specimen and performed the analysis considering plasticity of adhesives. Sawa et al. [9] researched on the effect of stiffness, yield stress of adherends, and joint length on the joint strength using finite element analysis. Schoeppner et al. [10] performed the progressive failure analysis of a double-lap joint using variational method and validated the analysis by agreement to experimental results. Takayuki et al. [11] compared the bond strength of double-lap joints composed of adhesives with different curing temperatures at low temperature. Weitsman [12] conducted an analysis of residual thermal stress considering elastic and viscoelastic properties of adhesive, respectively, using variational method when a double-lap joint undergoes temperature change. Roy et al. [13] analyzed the effect of moisture and temperature on the bond strength of a double-lap joint. Rastogi et al. [14] performed finite element analysis to obtain 3 dimensional thermal stress field of double-lap joint composed of aluminum and composite as adherends and certified that corner of joint part is the crack initiation point. However, in most cases, analyses and experiments were conducted at room temperature and there is little research on the bond strength at cryogenic temperature.

Therefore in this paper, 3 types of adhesives were selected. These are all film types. Graphite/epoxy composites developed for cryogenic use and aluminum for a lining material of the tank were bonded to make double-lap joint specimens with selected adhesives. Using an environmental chamber, tensile test of the double-lap joint specimens was conducted at room temperature and cryogenic temperature (-150°C) respectively to compare the bond strength of each adhesive and fracture characteristics. The material properties at various temperatures of component materials of the double-lap joint were measured. Also ABAQUS was used for the purpose of analyzing the experimental results.

2. EXPERIMENTAL

2.1 Materials used and double-lap joint specimen

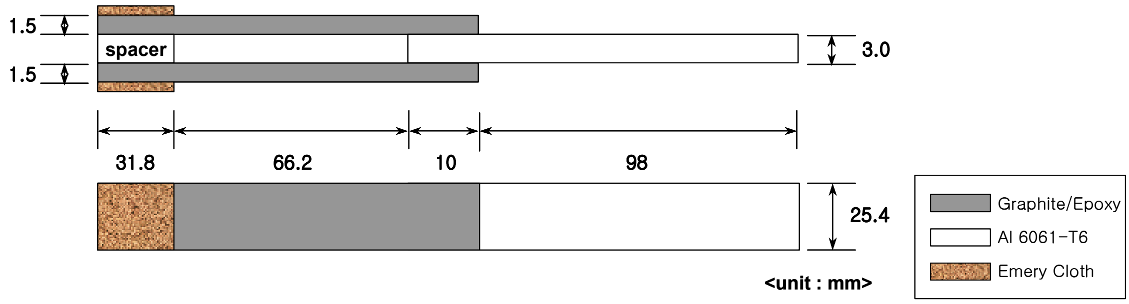


Fig. 1 Geometry of double-lap joint specimen

Double-lap joint specimens were manufactured as shown in Fig. 1 referring to ASTM D 3528. The composite part of the specimen is Graphite/Epoxy specially developed for cryogenic use and the reinforcing fibers are aligned and stacked along the length of the specimen. Aluminum alloy, Al 6061-T6, is the material generally used as a metal liner. The joint length is 10mm. After the joint surfaces are sanded using emery paper, with all other manufacturing conditions constant, the adhesive was applied. Also, the part of the composite where the grip is bitten was bonded with emery cloth for the tensile test under cryogenic environment. A spacer is used to maintain the distance between outer adherends and to protect the bending stress induced by the difference in thermal deformations of the spacer and the inner adherend.

The selected 3 kinds of adhesives between outer adherend composite and inner adherend aluminum are all film type and as follows: Bondex606 fabricated by Han-Kuk Fiber Glass Corporation, Korea, EA9696 by Loctite, FM73 by Cyanamid. The curing temperature of these 3 adhesives is 130°C same as that of the composite considered for actual fabrication of a tank.

2.2 Tensile test of double-lap joint specimens for bond strength at RT and CT

Three kinds of double-lap joint specimens after adhesive curing were used to perform tensile test and the bond strengths at RT (room temperature) and CT (cryogenic temperature, -150℃) were compared. As shown in Fig. 2, an environmental chamber was used to simulate RT and CT and 6 specimens were tested to obtain the bond strength at each temperature. General wedge type grips were used to perform the tensile test and the ram actuator was controlled at a rate of 1.27mm/min referring to ASTM D 3528. To prevent a specimen from sliding out of grips when being tensioned at CT, preload as much as 4kN was applied and the tensile test started after thermal equilibrium of the double-lap joint specimen was reached at CT.

Fig. 2 Tensile test of double-lap joint specimen in environmental chamber

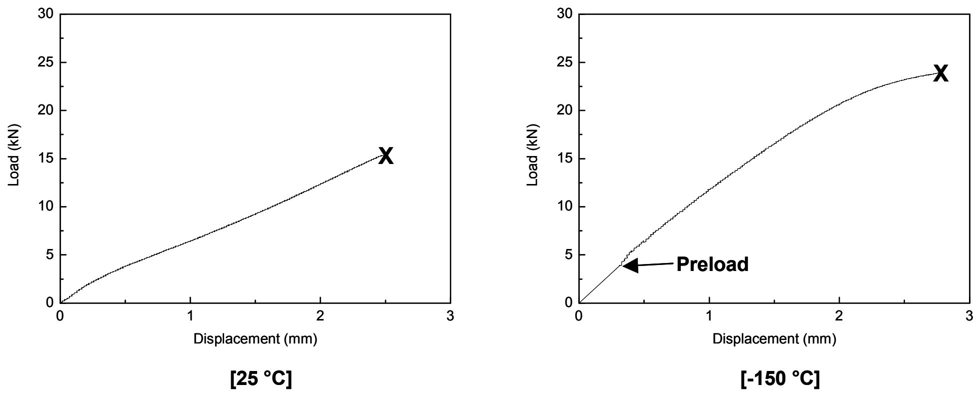


Fig. 3 Typical load-displacement curve of double-lap joint at RT and CT

Typical load-displacement curves of a double-lap joint at RT and CT are shown in Fig. 3. Obviously, the bond strength at CT is higher than that at RT. Comparison of bond strength of each adhesive is stated in section 3.1. Also in section 3.2, fracture characteristics of double-lap joints are observed and classified according to fracture modes.

2.3 Measurement of material properties of each component material of the double-lap joint at low temperature

The component materials that make up double-lap joint specimens, Graphite/Epoxy, three kinds of adhesives, and aluminum alloy, were tested to get their tensile stiffness, strength, and coefficient of thermal expansion at low temperature. Specimen preparation and tests for Graphite/Epoxy were conducted referring to ASTM D 3039. Also, bulk adhesive specimens were manufactured as dog-bone shape and tested referring to ASTM D 638, the aluminum alloy to ASTM B 557, respectively as shown in Fig. 4. Three types of bulk adhesive specimens can be discriminated apparently. The color of FM73 is brown, Bondex606 ivory and EA9696 bluish green. The thickness of these bulk adhesive specimens is 2mm more or less.

Using an environmental chamber, prepared specimens were tensioned at RT, -50°C, -100°C and -150°C respectively and the change of stiffness and strength was observed as temperature decreases. Also, a strain gage with half bridge circuit on titanium silicate was adopted as a dummy gage to get thermal strain and in turn, coefficient of thermal expansion at each temperature. These measured material properties are used to perform the finite element analysis of a double-lap joint specimen to get the stress distribution of the joint part.

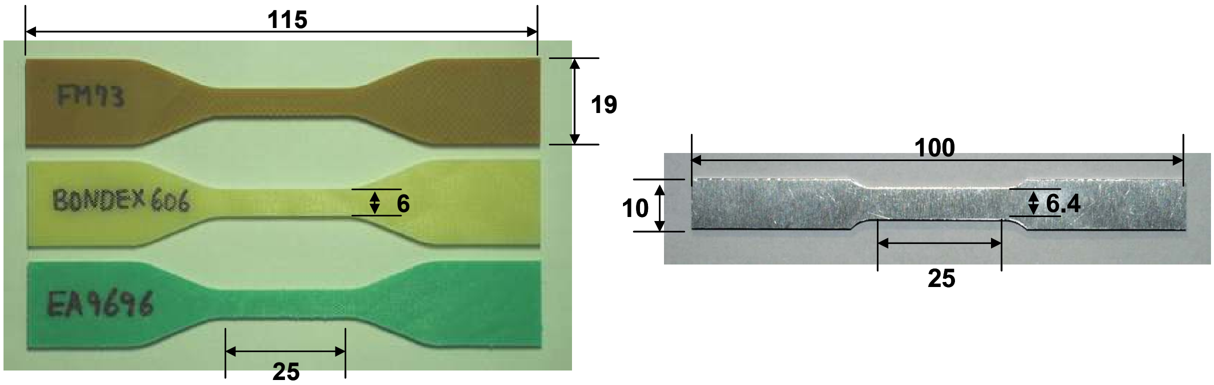


Fig. 4 Bulk adhesive specimens and an aluminum specimen

3. DISCUSSION

3.1 Experimental results of double-lap joint specimens

Experimental results of the 3 types of double-lap joint specimens are shown in Fig. 5 and numerical values are listed in Table 1. At room temperature, FM73 and Bondex606 have similar bond strength but EA9696 has shown low bond strength. At -150°C, bond strengths of the 3 types of double-lap joint specimens increase and Bondex606 has shown the highest bond strength among these. Comparing with the result of room temperature, FM73 shows an increase by 44.3%, Bondex606 by 53.0% and EA9696 by 89.8%. These phenomena seem to be led by the increase in strength of adhesive at CT and this can be verified from the experimental results of adhesives stated in section 3.3.

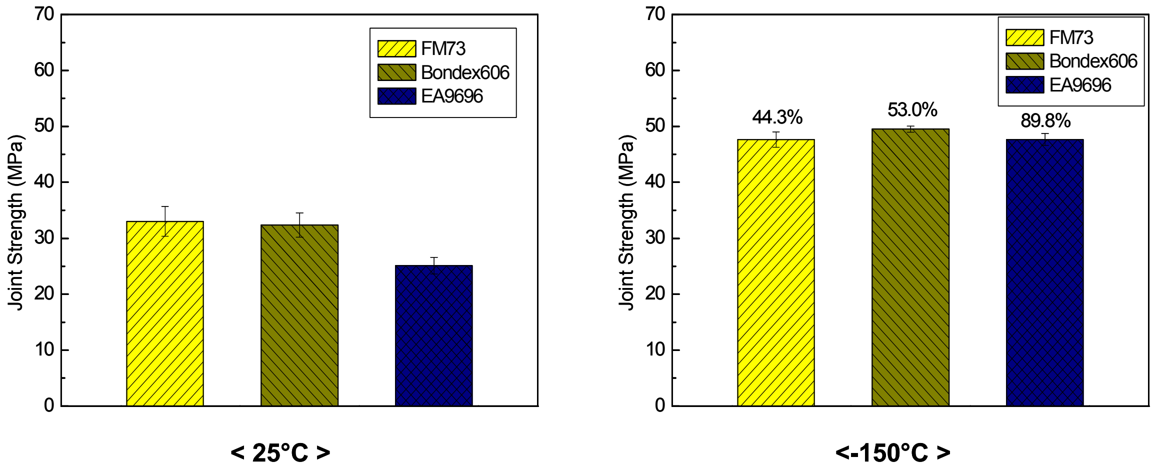


Fig. 5 Experimental results of double-lap joint specimens at RT and CT

Table 1 Experimental results of double-lap joint specimens at RT and CT

	FM73	Bondex606	EA9696
25°C	33.0 ± 2.64	32.4 ± 2.19	25.1 ± 1.47
-150°C	47.7 ± 1.36	49.5 ± 0.55	47.7 ± 1.06

(unit : MPa)

Also, it can be seen from the experimental results that adhesive, which has shown the most excellent bond strength at RT, doesn't always guarantee the excellence at CT like the case of FM73. Therefore, it must be preceded to understand the bonding characteristics of adhesives at low temperature before applying them at such an environment.

3.2 Fracture mode and fracture procedure of double-lap joint specimens

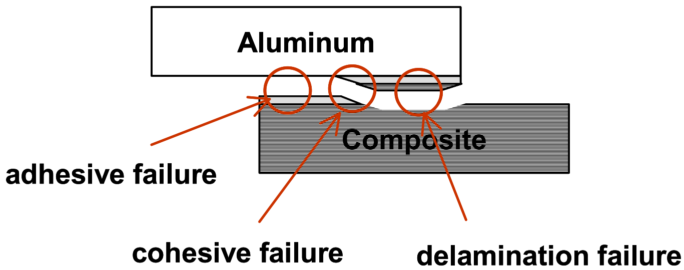


Fig. 6 Typical fracture mode of double-lap joint specimens

In Fig. 6, 3 typical fracture modes of a double-lap joint specimen (adhesive failure, cohesive failure, and delamination failure) are shown. The first means the failure caused by separation of adhesives from adherend clearly. The second is the failure in adhesive layer and the last in composite layer. In Fig. 7 fracture surface of FM73-inserted double-lap joint specimens at RT and CT can be seen, from which 3 types of fracture modes are observed. The three types of double-lap joint with 3 different adhesives have little difference in fracture modes both at RT and CT. However, at CT, the wreck of adhesive layer can be seen a little more than at RT.

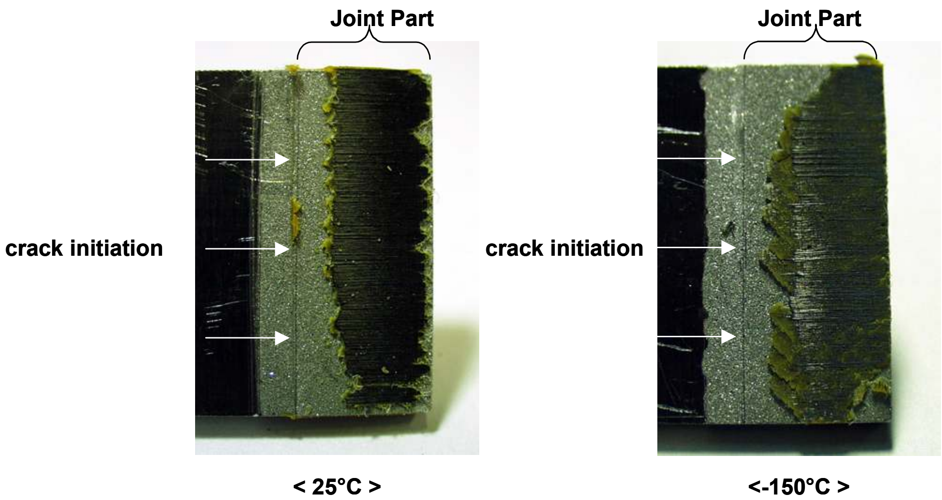


Fig. 7 Fracture surface of FM73-inserted double-lap joint specimen at RT and CT

In Fig. 7, the fracture procedure can be induced as follows. First, crack initiates at the joint part between the aluminum adherend and the adhesive layer (from left side in Fig. 7, adhesive failure) and propagates across the adhesive layer (cohesive failure). Finally, the crack propagates to the surface ply of the composite adherend and then that ply is separated with reinforcing fibers. That is the final failure of double-lap joint specimens.

3.3 Measurement of material properties of each component material of the double-lap joint at low temperature

Table 2 Material properties of Graphite/Epoxy at RT and CT

	E_1(GPa)	S_1(MPa)	E_2(GPa)	S_2(MPa)	G_{12}(GPa)	S_{12}(MPa)	α_1(με/°C)	α_2(με/°C)
RT	143.6	2930	8.87	53.7	4.50	65.1	-1.19	26
-150°C	161.3	2869	11.9	66.5	74.9	131.9	1.46	17.6

Material properties of Graphite/Epoxy at RT and CT are shown in Table 2. At CT, stiffness and strength tend to increase as a whole. This result reflects the increase in the properties of constituent reinforcing fibers and matrix at CT. It is noticeable that the coefficients of thermal expansion along reinforcing fibers have negative value at RT and positive at CT.

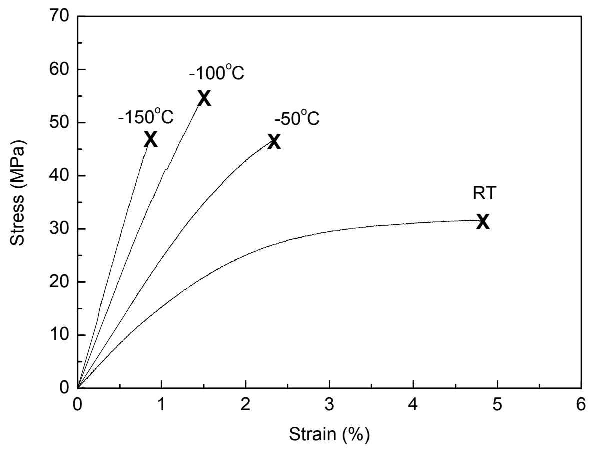


Fig. 8 Typical stress-strain curve of bulk adhesives at different temperature (EA9696)

Table 3 Stiffness and strength of 3 adhesives and aluminum at various temperatures

(a) Stiffness

	RT	-50°C	-100°C	-150°C
Bondex606	2.8	3.6	5.0	6.3
EA9696	1.9	2.4	3.9	5.5
FM73	2.5	3.2	4.5	5.5
Aluminum	68.5	70.6	72.0	74.1

(unit : GPa)

(b) Strength

	RT	-50°C	-100°C	-150°C
Bondex606	39.4	47.8	43.1	49.3
EA9696	33.9	50.0	53.8	46.4
FM73	45.7	73.3	54.0	57.4
Aluminum	294.6	313.5	321.3	350.5

(unit : MPa)

Typical stress-strain curve of EA9696 bulk adhesive at different temperature is shown in Fig. 8. As the temperature decreases, the slope of the stress-strain curve increases, from which the increase in the brittleness of adhesives brings about the increase in stiffness. Strength, also, increases comparing with the result at room temperature and this is the main reason of bond strength increase of double-lap joint specimens at CT.

In Table 3, stiffness and strength of 3 adhesives and the aluminum alloy, Al 6061-T6 is listed at various temperatures, where the strength of aluminum means the yield strength. Among the 3 adhesives, Bondex606 shows the highest stiffness and FM73 the highest strength over the whole temperature range. In Fig. 9, the measured coefficients of thermal expansion of the 3 adhesives and aluminum are shown and EA9696 has the highest value at every temperature.

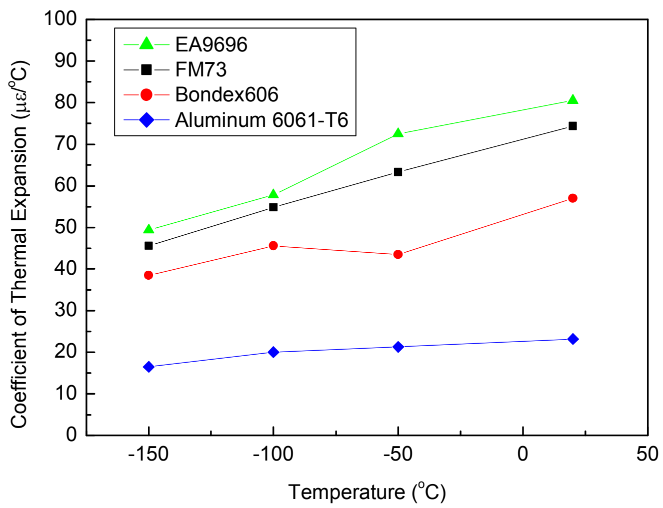


Fig. 9 Coefficients of thermal expansion of 3 adhesives and aluminum

3.4 Comparison of double-lap joint strength and bulk adhesive strength

Fig. 10 shows the comparison of double-lap joint strengths and bulk adhesive strengths at RT and CT. At room temperature, the double-lap joint strength doesn't come up to bulk adhesive strength. On the contrary, at -150°C, except FM73, double-lap joint strength and bulk adhesive strength have similar strength. Therefore, the excellence in the strength of a bulk adhesive specimen doesn't always mean that the joint strength is also excellent. And before applying adhesives to structure, adhesive test must be done in advance because bonding characteristic of adhesive can be varied according to service temperature and type of adherends.

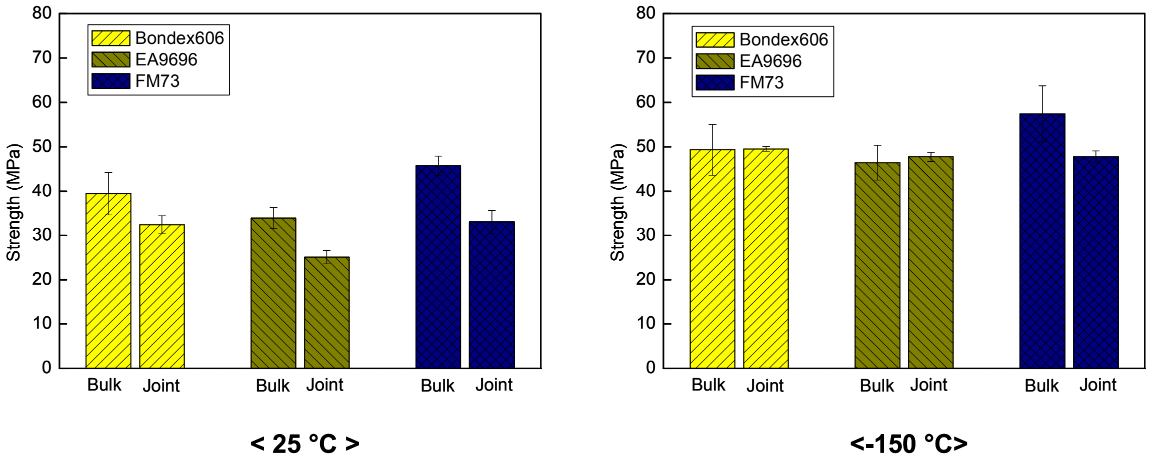


Fig. 10 Comparison of double-lap joint strength and bulk adhesive strength at RT and CT

4. FINITE ELEMENT ANALYSIS

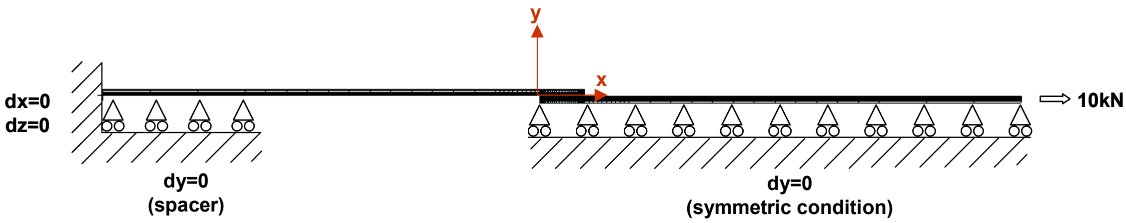
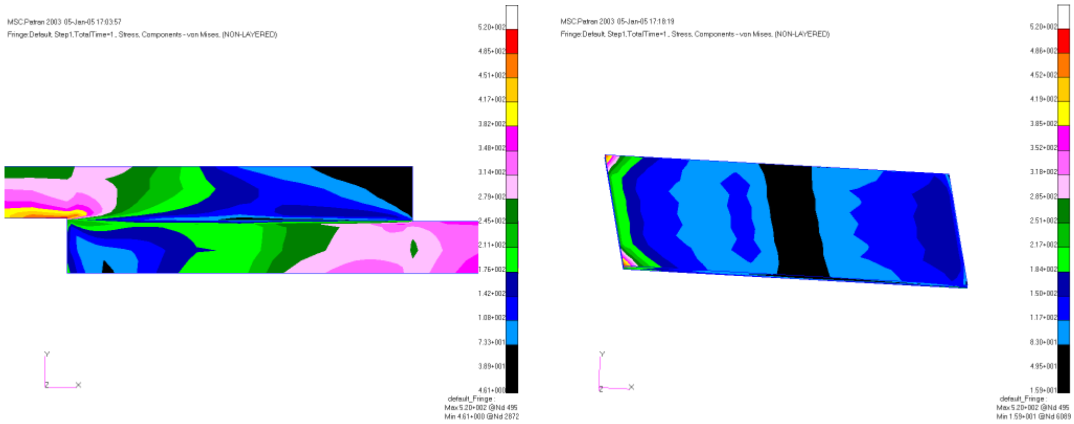


Fig. 11 Half model of double-lap joint and boundary condition for FEA

To analyze the stress field of the joint part of double-lap joint at -150°C, 3 dimensional finite element analysis was performed. MSC/PATRAN 2003 was used as a tool for pre processing including modeling, meshing, boundary condition, etc and post processing including displaying stress and strain results. And structural analysis was done by means of ABAQUS 6.4.

As shown in Fig. 11, for the half model of a double-lap joint, the boundary conditions and the loading were applied. Considering the spacer and its role, y-displacement is restricted in composite part adjacent to the spacer. And the symmetry condition was applied to the lower face of the inner adherend, aluminum alloy because it is cut along the centerline of the thickness of aluminum. Initial temperature was 20°C and -150°C was applied to all the nodes. And at the right face of the aluminum, a mechanical load of 10kN was applied.

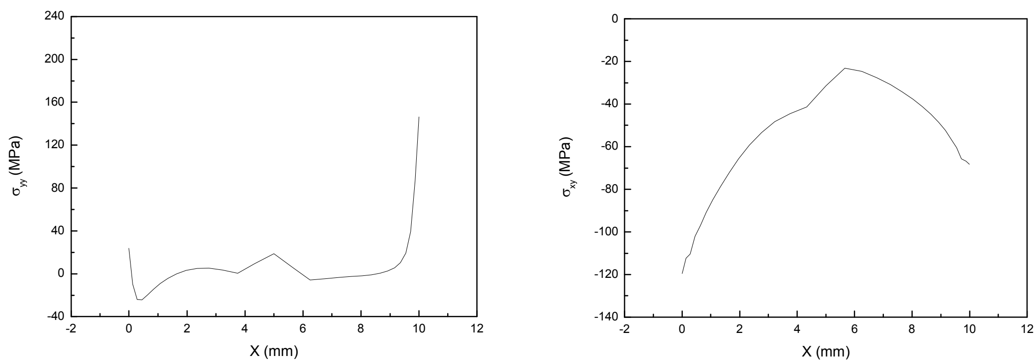
Eight-node solid elements were selected and the total number of nodes was 11429 and element 9400. Experimental data of each component material of a double-lap joint was input as material properties for the structural analysis. FM73 was used as an adhesive material in this analysis.



(a) Joint Part (b) Adhesive layer

Fig. 12 Von-Mises stress distribution of joint part and adhesive layer

In Fig. 12, von-Mises stress distribution of the joint part and the adhesive layer is shown. Complicated stress gradient of adherends can be seen from minimum 5MPa to maximum 500MPa. Especially, two edges of adhesive layer (x=0 and x=10) have quite high stress and this means partial failure or local plastic region of adhesive. In Fig. 13, at x=0 where adhesive layer comes in touch with free face of aluminum (left corner in Fig. 12 (a)), absolute value of shear stress, σ_{xy} , is high but peel stress, σ_{yy}, which is related to the crack opening and propagation, is low. On the contrary, at x=10 (right corner in Fig. 12 (a)), the absolute value of σ_{xy} is low but σ_{yy} is much higher than that at x=0. This can explain that the crack initiates at x=10 where peel stress is the highest over the joint length. This agrees to the fracture procedure of a double-lap joint in section 3.2 and the results of other papers [9-10].



(a) σ_{yy} (b) σ_{xy}

Fig. 13 Stress distribution (σ_{yy} and σ_{xy}) of adhesive layer

5. CONCLUSION

In this paper, 3 film types of adhesives were selected. Using these adhesives, 3 types of the double-lap joint specimens consisting of graphite/epoxy composites and aluminum were fabricated. And tensile tests of these specimens were conducted at room temperature and cryogenic temperature respectively to compare the bond strength of each adhesive. From the experimental results, Bondex606 has the highest joint strength at CT. And it can be seen the excellence of strength of bulk adhesive specimen doesn't always mean that of the joint strength. Therefore, the selection of adhesives for extreme temperature requires the adhesive test at operating temperature range in advance.

The material properties of component materials of double-lap joints at various temperatures were measured. Using these material data, ABAQUS was used for the purpose of analyzing the experimental results. The finite element analysis result can explain that fracture process.

6. REFERENCE

1. G. Vendroux, M. Auberon and J. Dessaut, 42nd international SAMPE Symposium, (1997)
2. R. Heydenreich, Cryogenics, 38, 125, (1998)
3. A. Pasquier, V. Peypoudat and Y. Prel, 23rd International Symposium on Space Technology and Science, (2002)
4. B. W. Grimsley, R. J. Cano, N. J. Johnson, A. C. Loos and W. M. McMahon, International SAMPE Technical Conference Series, 33, 1224, (2001)
5. K. S. Whitley and T. S. Gates, AIAA, (2002)
6. T. Shimoda, Y. Morino, T. Ishikawa, T. Morimoto and S. Cantoni, Proceeding of the Japan International SAMPE Symposium, 7, 275, (2001)
7. J. He, T. Shimoda, Y. Morino, and S. Mizutani, 23rd International Symposium on Space Technology and Science, (2002)
8. L. Tong, J. of Reinforced Plastics and Composites, 16, (8), 698, (1997)
9. T. Sawa and H. Suga, J. of Adhesion Science and Technology, 10, (12), 1255, (1996)
10. G. A. Schoeppner, A. K .Roy and S. L. Donaldson, AIAA, 1594, (1998)
11. T. Shimoda and J. He, 24th International Symposium on Space Technology and Science, (2004)
12. Y. Weitsman, J. of Thermal Stresses, 3, (4), 521, (1980)
13. A. K. Roy, S. L. Donaldson, Adv. mat.: develop., characterization process, and mechanical behavior, 73, (1996)
14. N. Rastogi, S. R. Soni and A. Nagar, Advances in Engineering Software, 29, (3-6), 273, (1998)
15. M. G. Kim, S. G. Kang, C. G. Kim, C. W. Kong, J.of Korean Society of Composite Materials, 17, (6), 52, (2004)
16. K. S. Kim, J. S. Park, Y. S. Jang and Y. M. Yi, J. of Korean Society of Composite Materials, 16, (5), 45, (2003)
17. ABAQUS/Standard User's Manual, Hibbitt, Karlsson & Sorensen, Inc.