DEVELOPMENT OF POLYIMIDE ADHESIVE FILM AND THE APPLICATION TO HIGH TEMPERATURE SANDWICH PANELS

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Abstract

Sandwich panels made of CFRP are widely used for structural components because of their excellent characteristics. Heat resistant sandwich panels will be required future aerospace systems operated in high temperature environment. The objective of this study is to develop a high temperature sandwich panel of which operating temperature exceeds 250 ºC. A novel phenylethynyl-terminated polyimide developed in JAXA was applied for the matrix resin of CFRP and film adhesive. The carbon foam was used for the core material. As a result, sandwich panels made of polyimide CFRP and carbon foam core were successfully fabricated. Mechanical properties of sandwich panels were evaluated using flatwise-tensile tests and three-point flexural tests. It was demonstrated CF/polyimide sandwich panels exhibit more excellent mechanical properties at 250 ºC as compared with CF/epoxy sandwich panels.

1. Introduction

A sandwich panel generally consists of two skin panels, lightweight core material, and adhesives. Sandwich panels are lightweight, and have high flexural stiffness. In aerospace field, sandwich panels made of carbon fiber reinforced plastic composites (CFRP) are widely used for structural components of aircrafts, satellites, and space transportation vehicles. The maximum operation temperature of current CFRP (CF/epoxy) sandwich panels is approximately 150 ºC for a short duration because of the degradation of matrix resin (epoxy) above 100 ºC. In the future, heat resistant sandwich panels which can be used at 200~300 ºC will be required for future aerospace systems operated at high temperatures such as Mars explorer, supersonic aircraft, hypersonic transportation systems, re-entry space vehicle. However, there are few reports of practical examples of heat resistant sandwich panels [1]. The heat resistance of skin panels, core materials, and adhesives is essential for the development of heat resistant sandwich panels. One of the promising core materials is carbon foam because of their excellent characteristics such as heat resistance with affordable cost [2]. In addition, candidate resin materials for the matrix of CFRP skin panels and adhesives are bismaleimide, benzoxazine, polyimide and so on. Among them, a novel phenylethynyl-terminated polyimide based on KAPTON backbone structure “TriA-X”, which was developed in Japan aerospace exploration agency (JAXA), is applied in this study [3, 4]. TriA-X exhibits both excellent moldability and heat resistance of 350 ºC or more. Additionally, because this resin does not produced by-products during curing, this resin is expected as the thermosetting adhesive.

The objective of this study is to establish the processing of a high temperature sandwich panel of which operating temperature exceeds 250 ºC. Heat resistant film adhesive using TriA-X imide oligomer was developed. Carbon/polyimide-composite/carbon-foam-core sandwich panels were fabricated using the film adhesives. The mechanical properties of sandwich panels were examined using flatwise-tensile tests and three-point flexural tests. The effects of thermal residual stress on the strength of sandwich panels were also evaluated using finite element analyses.

2. Experimental procedures

2.1. Processing of imide oligomer film adhesives

Film adhesive was fabricated impregnating TriA-X imide oligomer (n=6) into glass fiber mesh cloth (E03A, UNITIKA LTD). The chemical structure of TriA-X imide oligomer is shown in Figure 1. Imide oligomer/NMP (N-methylpyrrolidone) solution (35 wt.% of imide oligomer) was uniformly spread on a UPILEX polyimide film (thickness 0.075 mm) on a glass plate using a film applicator. A glass fiber mesh cloth (areal weight of 24 g/m^2) was put on it, and the imide oligomer/NMP solution was impregnated into the glass cloth. The same procedure was carried out for the back side of the same glass fiber cloth. The solvent (NMP) was evaporated in an oven. The temperature was increased from 50 ºC to 250 ºC with step-wise increment of 50 °C. The holding time for each step was 30 minutes.

Three kinds of film adhesives, namely 75-75, 100-100, and 125-125, were made to investigate the effect of adhesive thickness on mechanical property. The 75-75, 100-100, and 125-125 denote the thickness (in micrometer) of imide oligomer/NMP solution spread using a film applicator. After removing NMP, the areal weight of the film adhesives is 65 g/m^2, 117 g/m^2, and 172 g/m^2 for 75-75, 100-100, and 125-125 adhesives respectively.

2.2. Single lap shear test

The single lap shear test was performed in order to evaluate the mechanical property of the TriA-X film adhesive. Schematic drawings of a jig and a specimen are shown in Figure 2. Plain-woven carbon fabric (IMS60, Toho Tenax, Japan) /TriA-X composite laminates (16 ply, 12 mm length, 8 mm width, 2.6 mm thickness) were prepared for adherend. Two composite laminate specimens were bonded together at 310 ºC for 15 minutes and at 370 º C for 60 minutes under the pressure of about 0.1 MPa. The adhesion area was 8 mm width and 5 mm length.

The single lap shear tests were performed at 25 ºC under a constant crosshead speed of 1 mm/min. For 125-125 adhesive, single lap shear tests were also carried out at 200 ºC, 250 ºC, and 300 ºC. The shear strength $\boldsymbol{\tau^f}$ was calculated using the following equation (1).

$$\tau^f = \frac{P_{\max}}{A} \tag{1}$$

$\boldsymbol{P_{\max}}$: **maximum load**, $\boldsymbol{A}$: **adhesion area**

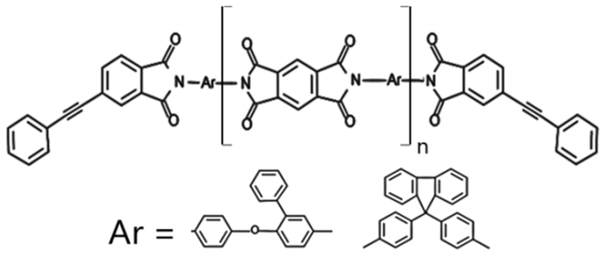


Figure 1. Chemical structure of TriA-X imide oligomer.

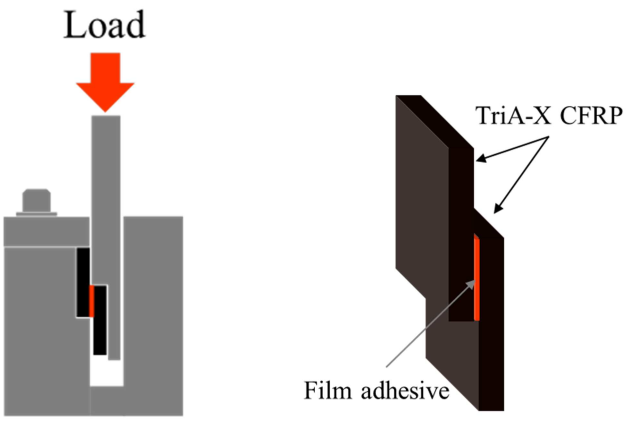


Figure 2. Schematics of jig and specimen for the single lap shear test.

2.3. Processing of sandwich panels

Plain-woven carbon fabric (IMS60, Toho Tenax)/ TriA-X polyimide composite laminates (3 ply, 0.6 mm thickness) were prepared for skin panels of sandwich panels. Porous carbon foam (Grafoam FPA-10, density 0.16 g/cc, GrafTech) was used for the core material. 125-125 film adhesive was used because this exhibited the highest strength at 25ºC. The skin panels were bonded on the carbon foam core at 310 ºC for 15 minutes and at 370 º C for 60 minutes under the pressure of about 0.1 MPa. To improve the bonding strength, the TriA-X imide oligomer was previously dispersed at the surface of carbon foam core. The procedure was the same to make film adhesives, and the thickness of spread imide oligomer/NMP solution was 100 μm, and 250μm.

For direct comparison, CF (IMS60, Toho Tenax, $[0/90/\pm45]_s$)/epoxy sandwich panels were fabricated using epoxy film adhesive (AF163, 3M). The fabrication process of CF/epoxy sandwich panels was to hold 60 minutes at 122 ºC under the pressure of about 0.1 MPa.

2.4. Test method for sandwich panels

The mechanical properties of sandwich panels were evaluated using flatwise-tensile tests according to ASTM C297, and three-point flexural tests according to ASTM C393. The specimen size of flatwise-tensile tests was 25mm×25mm×25mm, and the test configuration is shown in Figure 3. Two loading blocks made of aluminum alloy were bonded to top and bottom sides of a specimen using epoxy adhesive (Araldite, Huntsman Advanced Materials). The crosshead speed was 0.5 mm/min. The effect of imide-oligomer pre-impregnated thickness to the carbon foam surface on the flatwise-tensile strength was evaluated. The flatwise-tensile strength $\boldsymbol{F_Z}$ was calculated using the following equation (2).

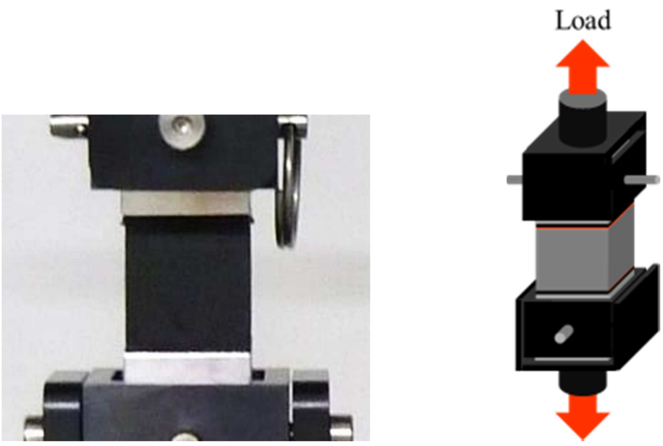


Figure 3. Test configuration for the flatwise-tensile test.

$$F_Z = \frac{P_{\max}}{A} \tag{2}$$

$\boldsymbol{P_{max}}$: **maximum load,** $\boldsymbol{A}$**: adhesion area**

A three-point flexural test specimen was 120 mm in length, and 25 mm in width. The thickness of CF/TriA-X sandwich panels was 6 mm, whereas that of CF/epoxy sandwich panels was 7.5 mm. The lower span length was 100 mm. Flexural tests were conducted at 25 ºC, 200 ºC, 250 ºC, and 300 ºC. To prevent the failure at the loading point, silicone rubber sheet was put between a specimen and each loading roller. The test configuration for the three-point flexural test is shown in Figure 4. For three-point flexural tests of sandwich panels, the shear fracture at skin/core interlayer occurred for all of the specimens. Therefore experimental results are discussed using shear stress at the skin/core interface layer. The shear strength $\boldsymbol{\tau_b}$ was calculated using the following equation (3).

$$\tau_b = \frac{E_s P_{\max}}{16(E_s I_1 + E_c I_2)}\left(t^2 - {t_c}^2\right) \tag{3}$$

$$\boldsymbol{I_1 = 2w\int_{\frac{t_c}{2}}^{\frac{t}{2}} y^2\,dy,\quad I_2 = 2w\int_{0}^{\frac{t_c}{2}} y^2\,dy}$$

$\boldsymbol{P_{\max}}$: **maximum load,** $\boldsymbol{E_s}$**: young modulus of the skin panel**
$\boldsymbol{E_c}$: **young modulus of the core material,** $\boldsymbol{w}$**: width of the sandwich panel**
$\boldsymbol{t}$**: thickness of the sandwich panel,** $\boldsymbol{t_c}$**: thickness of the core material**

3. Result and discussion

3.1. Fabrication of adhesives and sandwich panels

Figure 5 shows a trial product of TriA-X film adhesive. Although the film adhesive has no tackiness, it is flexible and easy to cut to a desired shape. The cross-section of CF/TriA-X sandwich panel was inspected using an X-ray computed tomography scanner (X-CT). The cross-sectional image is shown in Figure 6. Some small voids are visible between adhesive and the core material. However they are the original pores of core material itself. The skin and core seem to adhere very well.

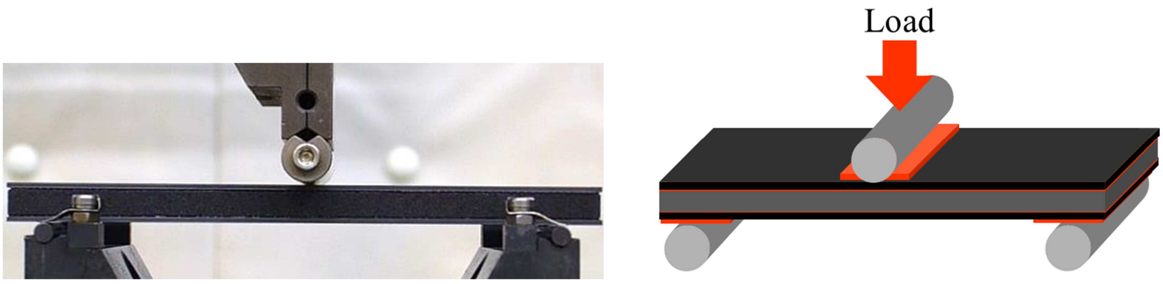


Figure 4. Test configuration for the three point bending test.

3.2 Single lap shear tests

The experimental results of single lap shear tests are summarized in Figure 7 for 75-75, 100-100, and125-125 specimens. The shear strength increases with adhesive thickness. It implies the sufficient amount of imide oligomer is necessary to keep high strength of the bonded joints. Figure 7 (b) shows the experimental results of single lap shear tests at high temperature. The shear strength is almost constant up to 200 ºC. Although the strength decreases gradually above 200 ºC, it is still about 70% of the strength at 25 ºC. The experimental results suggest that the TriA-X adhesive can be used above 200 ºC.

3.3 Flatwise-tensile tests

The experimental results of flatwise-tensile tests are presented in Figure 8. The effect of pre-impregnated imide oligomer depth on the tensile strength is not clearly observed. It suggests that skins and the core material is well-bonded using TriA-X film adhesive. The flatwise-tensile strength of TriA-X sandwich panels is about 60% of that of CF/epoxy sandwich panels at 25 ºC. Figure 9 (a) and (b) respectively show CF/epoxy and CF/TriA-X specimens after tensile testing. CF/epoxy sandwich panels were fractured at the core material, while CF/TriA-X sandwich panels were fractured near adhesive interface. It seems to be attributed to the residual thermal stress at the adhesive interface because of higher process temperature for polyimide adhesive (370 ºC) as compared with that for epoxy adhesive (122 ºC).

Figure 5. Trial product of TriA-X film adhesive.

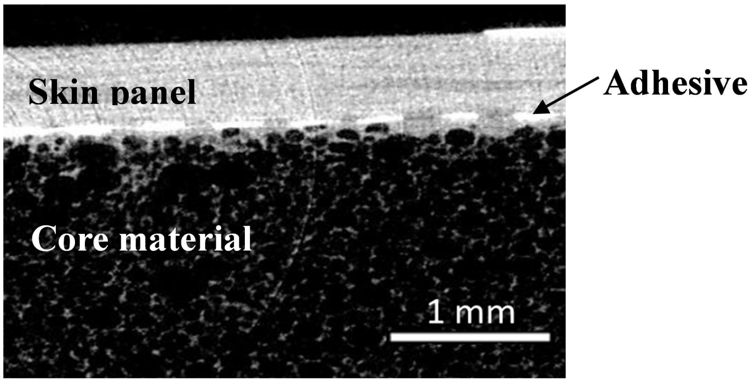
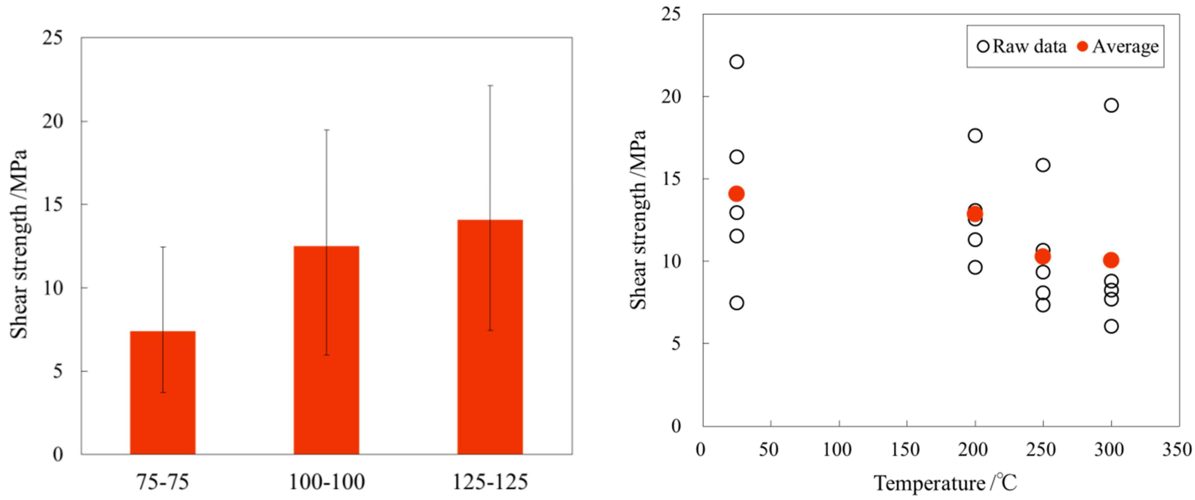


Figure 6. Cross-sectional image of CF/TriA-X sandwich panel.



(a) shear strength at 25 ºC (b) shear strength at high temperature

Figure 7. Shear strengths of TriA-X film adhesives.

3.4 Three-point flexural tests for sandwich beam

Figure 10 shows the specimen after flexural testing. The shear fracture at skin/core interlayer occurred for all of the specimens regardless of temperature [5]. Therefore the experimental results are discussed using shear stress at the skin/core interface layer as mentioned above. Three-point flexural test results are shown in Figure 11. For CF/epoxy sandwich panels, the shear strength at 200 ºC is degraded to be approximately a half of that at 25 ºC. On the other hand, the shear strength of CF/TriA-X sandwich panels is almost constant up to 250 ºC, and it is higher than that of CF/epoxy sandwich panels. Although the shear strength of CF/TriA-X sandwich decreases at 300 ºC, it is still 80% of the strength at 25 ºC. As results, it has been demonstrated that CF/TriA-X sandwich panels exhibit more excellent strength above 200 ºC as compared with CF/epoxy sandwich panels.

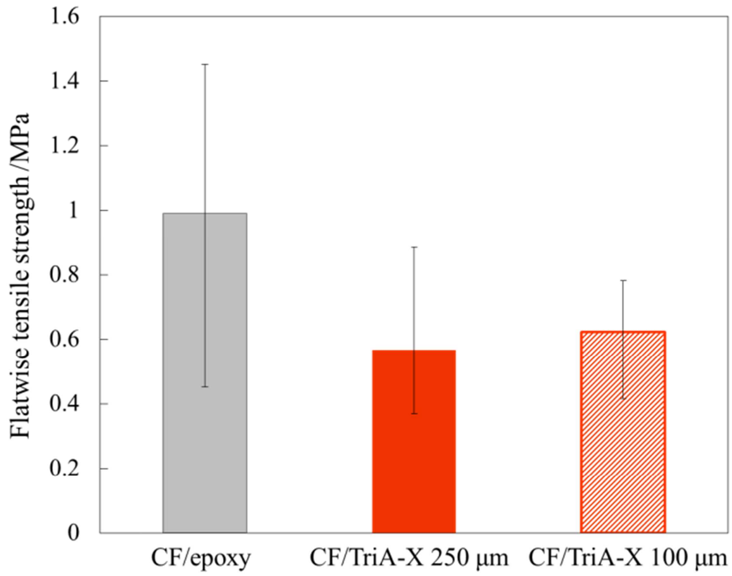
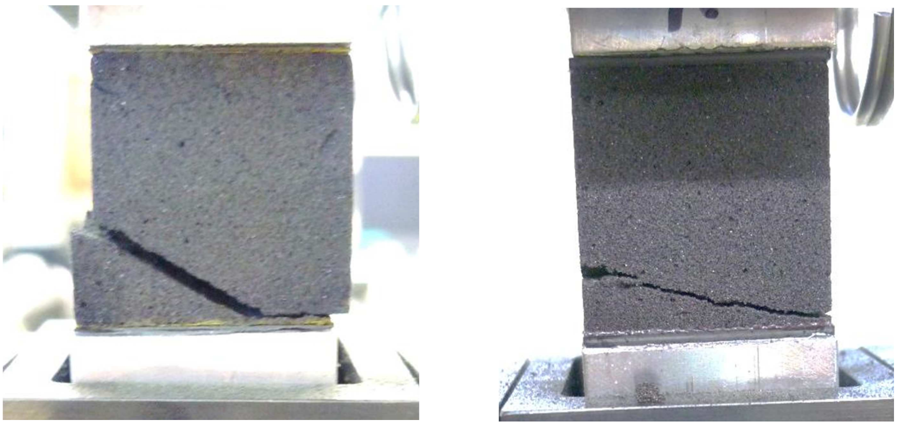


Figure 8. Flatwise-tensile strength of sandwich panels.



(a) CF/TriA-X sandwich panels (b)CF/epoxy sandwich panels

Figure 9. Flatwise-tensile specimens after tensile testing.

Figure 10. CF/TriA-X sandwich panel after flexural testing.

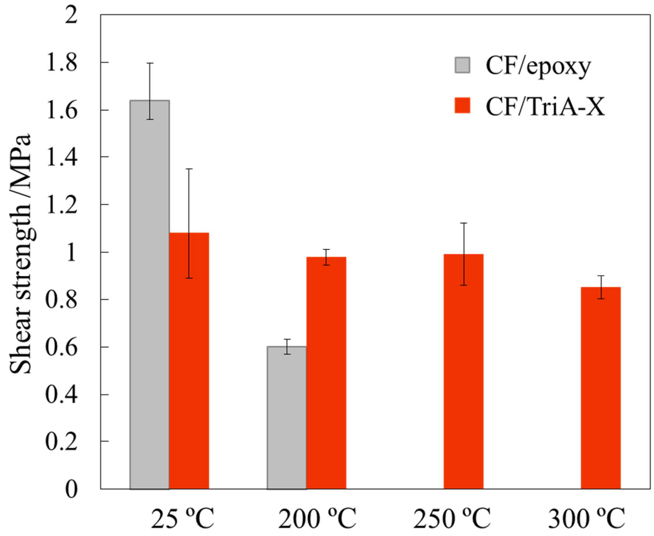


Figure 11. Shear strength of sandwich panels.

4. Conculusion

Imide-oligomer film adhesives were fabricated using a new phenylethynyl-terminated polyimide, TriA-X. Polyimide film adhesive exhibited high shear strength above 200 ºC. Carbon/polyimide-composite /carbon-foam-core sandwich panels were fabricated using the film adhesives. Cross-sectional inspection showed skin panels and the core material were bonded well. Although the flatwise-tensile and three-point flexural strengths of CF/polyimide sandwich panels were lower than those of CF/epoxy sandwich panels at room temperature, the strengths of CF/polyimide sandwich panels were almost constant up to 250 ºC. As results, it has been demonstrated the imide-oligomer film adhesives, and the CF/polyimide sandwich panels exhibit more excellent mechanical properties above 250ºC as compared with CF/epoxy sandwich panels.

References

[1] C. J. Camarda, Experimental investigation of graphite/polyimide sandwich panels in edgewise compression, NASA Technical Memorandum 81895 (1980).
[2] G. Reyes, S. Rangaraj, Fracture properties of high performance carbon foam sandwich structures, Composites: Part A, 42, 1~7, 2011.
[3] M. Miyauchi, Y. Ishida, T. Ogasawara, R. Yokota, Novel phenylethynyl-terminated PMDA-type polyimides based on KAPTON backbone structures derived from 2-phenyl-4,4′ -diaminodiphenyl ether, Polymer Journal (2012) 44, 959-965.
[4] Y. Ishida, M. Miyauchi, T. Ogasawara, R. Yokota, DEVELOPMENT OF “TRIA-X”POLYIMIDE/CARBON FIBER COMPOSITES PREPARED BY IMIDE SOLUTION PREPREGS”, Proceedings of 18th International Conference on Composite Materials, 2011
[5] M. D. Sarzynski and O. O. Ochoa, Carbon foam core composite sandwich beams: flexura response, Journal of Composite Materials (2005) 39 [12], 1067--1079.