MEASUREMENT OF THE MECHANICAL PROPERTIES OF A CARBON REINFORCED BISMALEIMIDE OVER A WIDE RANGE OF TEMPERATURES

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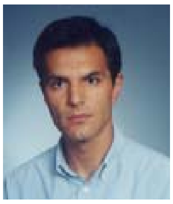

ABSTRACT

For high temperature usage (200°C and above) such as in certain supersonic aircraft structures, the composites used are either bismaleimides (BMI) or polyimides reinforced with carbon fibres. The composite selected was Advanced Composites Group HTM552. It is a carbon fabric 0/90° laminate of high strength carbon fibres in a 2 × 2 twill weave impregnated with a BMI resin. To determine the mechanical behaviour of the BMI composite as a function of temperature, the following tests were performed. The longitudinal E (E_{11}) was obtained dynamically by vibrating a free-free beam in flexure from –55 to 200°C. The ultimate longitudinal strength (σ_{11}) was determined using the 4-point flexure test. The through thickness strength (σ_{22}) was determined by bonding a piece of composite to steel blocks and loading in tension normal to the fibre direction. Finally, the coefficient of thermal expansion of the BMI composite was measured using strain gauges according to a method proposed by Lord.

1- INTRODUCTION

Adhesive joints used in supersonic aircraft need to withstand low (-55°C) when travelling subsonically at high altitude and high temperatures (200°C or so) when travelling at Mach 2 or above. Epoxies may not generally be used at high temperatures (over 150°C) although some may be used up to 200°C. For high temperature applications, the adhesives used are either bismaleimides or polyimides. Adhesives suitable for high temperatures are generally too brittle at low temperatures. Owing to their brittleness and high stiffness, the strength in a joint is poor at low temperatures. On the other hand, adhesives suitable for low temperatures are too weak or degrade at high temperatures. It may eventually be possible one day for chemists to develop an adhesive which will operate from -55°C to 200°C, but this is unlikely in the near future. One solution is to build a joint with a combination of two adhesives, consisting of a low temperature adhesive which is tough at low temperatures and has a high modulus, but which does not degrade at the highest required operating temperature even though it may be very ductile and creep, and a high temperature adhesive which has high strength at high temperatures but may be very brittle at low temperatures. The authors have previously shown that this technique enables to have a joint with a higher load capacity than a joint with only one adhesive, especially for dissimilar adherends **(da Silva and Adams, 2005)**.

The mixed adhesive joint is required to work when bonding composites to metal in the fuselage of supersonic aircraft. The metals commonly used are aluminium or titanium. The composites usually have a high-performance epoxy or BMI matrix. The range of temperature being studied is –55°C to 200°C. At 200°C, the aluminium starts to soften and the epoxy resins do not withstand well that temperature. Therefore, titanium was chosen for the metal and carbon reinforced BMI for the composite. The adherends were tested in-house.

The composite selected was Advanced Composites Group HTM552. It is a carbon fabric 0/90° laminate of high strength carbon fibres in a 2 × 2 twill weave impregnated with a BMI resin. The composite was purchased in the prepreg form and was manufactured in-house in an autoclave. The autoclave cure was at 190°C for 6 h. The postcure was done in a freestanding condition in an oven at 240°C for 6 h. 18 plies were arranged to give a 4 mm thick adherend. According to the manufacturer, T_g after cure is approximately 195°C and after postcure it is 282°C. This composite was developed for use in structures where critical load-bearing performance is required at temperatures greater than 200°C. The Young's modulus in the 0º and 90º directions (E_{11} and E_{33}) was obtained dynamically by vibrating a free-free beam in flexure from –55 to 200°C. The ultimate strength in the 0º and 90º directions (σ_{11} and σ_{33}) was determined using the 4-point flexure test **(ASTM D 790-71)**. The coefficient of thermal expansion (CTE) of the BMI composite in the 0º and 90º directions (CTE_{11} and CTE_{33}) were measured using strain gauges according to a method proposed by **Lord (1997)**.

2 – 0º AND 90º MODULI

The Young's modulus in the 0º and 90º directions was obtained dynamically by vibrating a small free-free beam in flexure at its first resonance frequency from –55 to 200°C. The frequency at resonance (f_n) of a plain beam is given by Equation 1:

$$f_n = \frac{1.027t\sqrt{\dfrac{E}{\rho}}}{l^2} \tag{1}$$

where t is the thickness (mm), E the Young's modulus (Pa), ρ the density (kg/m^3) and l the length (mm). An electrodynamic shaker was used to excite the beam and a laser to measure the resonance amplitude. The set-up used to find resonance is shown in Fig 1.

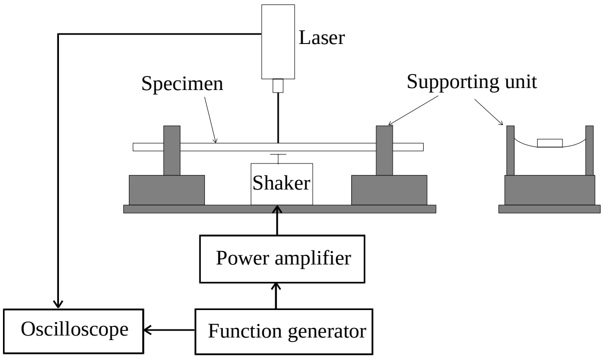

Fig 1 - Set-up for dynamic Young's modulus.

This method was used in preference to a static test since the latter is less accurate due to errors in calibration and measurement. The variation of E_{11} and E_{33} as a function of temperature is presented in Fig 2. Taking into account the CTE significantly affects the results. Neglecting the CTE, the modulus increases with temperature whereas with CTE it decreases.

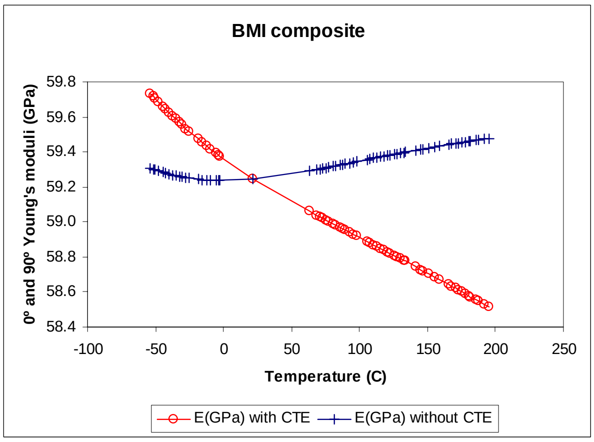

Fig 2 - 0º and 90º Young's moduli of the BMI composite.

Adams and Gaitonde (1993) tested dynamically unidirectional composites in the longitudinal direction at cryogenic temperatures and found similar results. The CTE in the width direction is the same as in the longitudinal direction due to the nature of the laminate and was found to be 4.2×10^{-6} °C^{-1}. (see section ‘Coefficient of thermal Expansion’). In the transverse (through the thickness) direction, the CTE can be estimated using Equation 2, proposed by Rojstaczer *et al* (1985) and referred to by Adams and Gaitonde (1993):

$$\mathrm{CTE}_{\mathrm{c}}^{\mathrm{t}} = \mathrm{CTE}_{\mathrm{ma}} V_{\mathrm{ma}} \left(1 + \nu_{\mathrm{ma}}\right) + \mathrm{CTE}_{\mathrm{f}}^{\mathrm{r}} V_{\mathrm{f}} \qquad (2)$$

where V is the volume fraction and ν the Poisson’s ratio. The subscript t stands for transverse, c for composite, ma for matrix, r for radial and f for fibre. The manufacturer gave a $\mathrm{CTE}_{\mathrm{ma}}$ of 45×10^{-6} °C^{-1}, a ν_{ma} of 0.35 and a $\mathrm{CTE}_{\mathrm{f}}^{\mathrm{r}}$ of 10×10^{-6} °C^{-1}. The volume fraction was estimated using the density measurement method (Curtis, 1988). The density of the matrix is 1210 kg/m^3 (manufacturer), the density of the carbon fibre is 1780 kg/m^3 (manufacturer) and the density of the composite was measured and found to be 1510 kg/m^3. This yielded a volume fraction of fibre of 53%. Equation 2 gives a $\mathrm{CTE}_{\mathrm{c}}^{\mathrm{t}}$ of 34.4×10^{-6} °C^{-1}.

Note that when working with an isotropic material such as titanium, the Young’s modulus is nearly not affected by the CTE as shown in **Fig 3**.

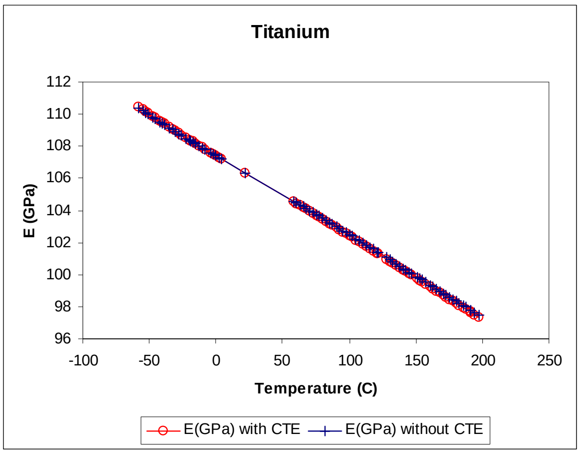

Fig 3 - Titanium Young’s modulus as a function of temperature.

3 - FOUR-POINT FLEXURE TEST (ASTM D 790-71)

The dimensions of the specimen together with the set-up are presented in Fig 4. The load-displacement curve was linear in all cases. The longitudinal strength as a function of temperature is given in Fig 5. Tests were performed at –55, 22, 100 and 200°C. It is interesting to note that the strength increases with temperature. The resin becomes tougher and more ductile as the temperature approaches T_{g} and is less sensitive to defects.

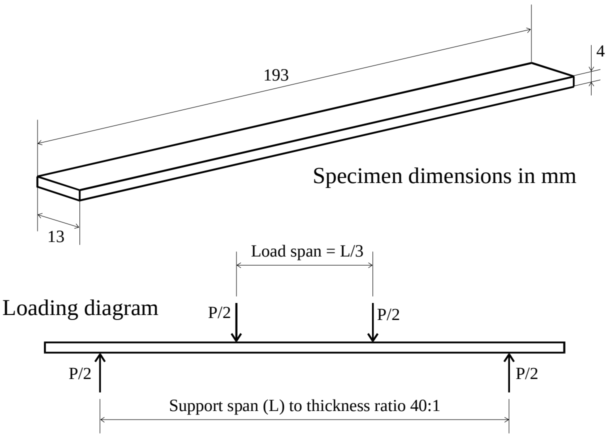

Fig 4 - Four-point flexure test set-up.

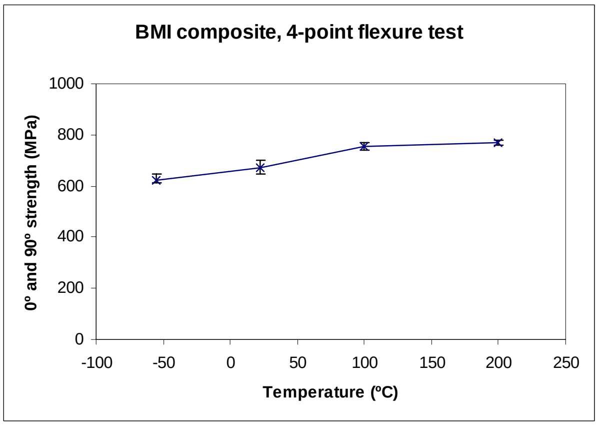

Fig 5 - 0° and 90° strength in flexure of the BMI composite.

4 - TRANSVERSE (THROUGH THE THICKNESS) STRENGTH

It is widely acknowledged that the through-thickness strength of composites is a difficult property to measure. In this study, a piece of the composite plate used to manufacture the double lap joints was cut and bonded to two steel blocks waisted to 12 × 15 mm. The specimen was then loaded normal to the fibre direction. It is necessary to guarantee the load is perfectly aligned and that there is no bending effect that would lead to a premature failure. This was achieved by loading the steel blocks through precisely aligned pins as shown in Fig 6.

Preliminary specimens where the glueline was very thin (0.1 mm) failed at the adhesive/composite interface. A finite element analysis (ABAQUS) was carried out in 2D (plain strain) to determine the stress distribution at the interface and to improve the initial design (see Fig 7a). It was thought first that the square edges of the composite introduced a high stress concentration. A chamfer was introduced in the composite as shown in Fig 7c. This reduced the stress concentration. However, the failure was still at the adhesive/composite interface, even though the failure load was higher. A third design was investigated where the glueline thickness is 1 mm. The stress distribution was now much more uniform (see Fig 7d) and the failure took place in the composite, very close to the adhesive. This design was adopted for the tests. A jig was used to guarantee alignment during manufacture. The adhesive used to bond the composite to the steel blocks was the epoxy AV119 from Vantico. Its T_g is 120°C so that the composite was not tested at 200°C. The results are presented in Figure 6. Contrary to the longitudinal strength, the transverse strength decreases with temperature although, at –55°C, the strength is slightly lower than at 22°C, probably because the resin is very brittle at that temperature. In general, the transverse strength of a composite is less than that of the unreinforced matrix because cracks form at the fibre/resin interface and link up through highly stresses sections of the matrix (Hull and Clyne, 1996).

5 - COEFFICIENT OF THERMAL EXPANSION

The coefficients of thermal expansion were measured using strain gauges according to a method proposed by Lord (1997).

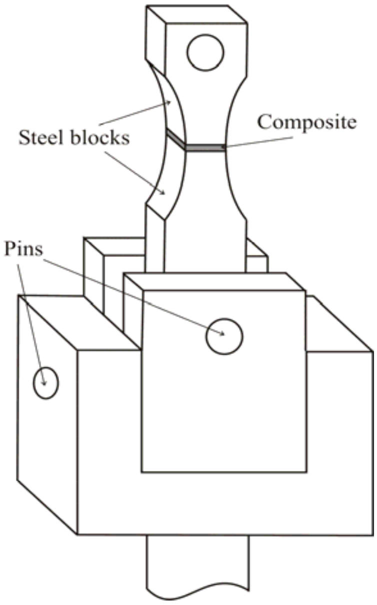

Fig 6 - Pin loading arrangement for the transverse strength specimen.

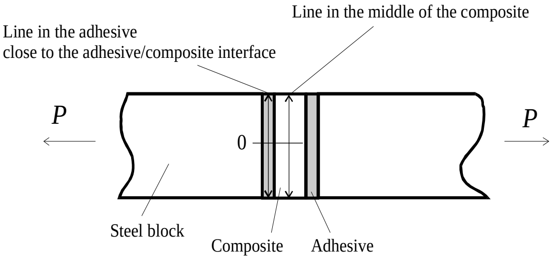

Fig 7a - Joint for the determination of the trough the thickness strength of the composite and location of the adhesive and composite stresses represented in Figures 7b, c and d.

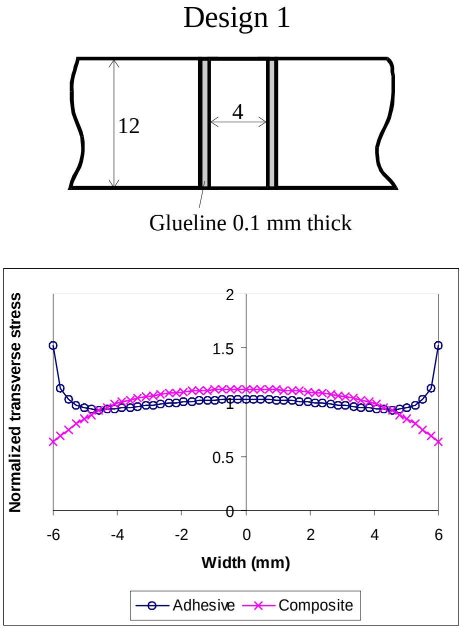

Fig 7b Normalized transverse stresses in the adhesive close to the composite and in the middle of the composite for design 1.

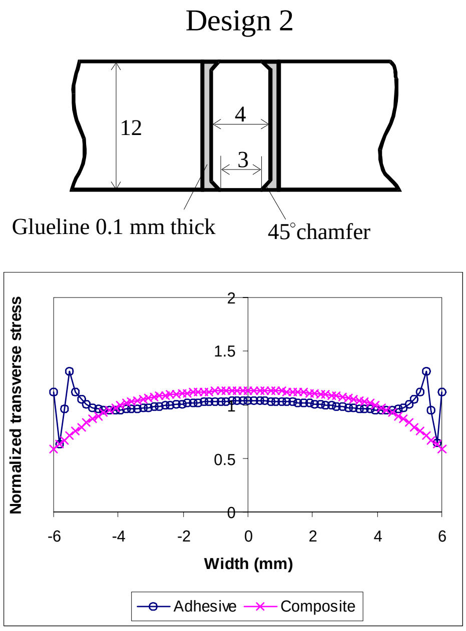

Fig 7c Normalized transverse stresses in the adhesive close to the composite and in the middle of the composite for design 2.

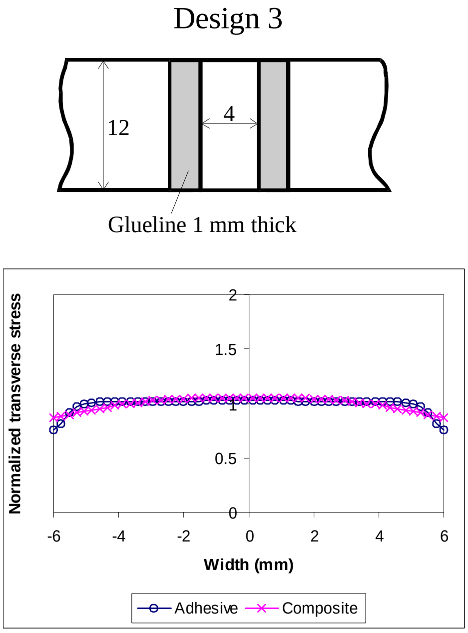

Fig 7d Normalized transverse stresses in the adhesive close to the composite and in the middle of the composite for design 3.

The technique uses two identical strain gauges, one of which is bonded to a specimen of a well-defined reference material, the second to the material of interest. Under stress-free conditions and at a common temperature, the differential output of the two gauges represents the difference in thermal expansion of the two materials. If the thermal expansion behaviour of the reference material is well characterised, the CTE of the material under examination can be calculated using the following equation:

$$\mathrm{CTE}_{\mathrm{spec}} = \frac{\left(\varepsilon_{\mathrm{spec}} - \varepsilon_{\mathrm{ref}}\right)}{\Delta T} + \mathrm{CTE}_{\mathrm{ref}} \qquad (3)$$

where $\mathrm{CTE}_{\mathrm{spec}}$ and $\mathrm{CTE}_{\mathrm{ref}}$ refer to the CTEs for the specimen and reference material over the temperature range ΔT and $\varepsilon_{\mathrm{spec}}$ and $\varepsilon_{\mathrm{ref}}$ are the corresponding strain values. The strain and the temperature were measured according to the experimental set-up represented in Fig 9. The reference material was the aluminium alloy 2024-T3. According to the literature, its CTE is 22.9×10^{-6} °C^{-1} between 20 and 100°C (Dieter, 1997). Using Equation 3, the CTE of the composite can be estimated. Figure 8 presents the raw data (microstrain vs. temperature). The experiment was carried out between the temperature of 25 and 60°C. To check that the technique is adequate, the CTE of mild steel was first measured. The value obtained was 11.7×10^{-6} °C^{-1} which is typical for this kind of material (Dieter, 1997). The CTE in the width direction is the same as in the longitudinal direction due to the nature of the laminate and was found to be 4.2×10^{-6} °C^{-1}. The CTE of carbon reinforced plastics is generally close to zero, but there can be variations from negative to positive values, depending on the fibre used, the fabric geometry, and the fibre volume fraction (Ganesh and Naik, 1994; Kim et al

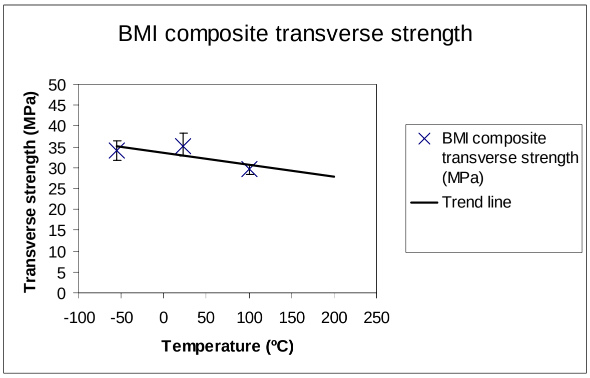

Fig 8 - Transverse (through the thickness) strength of the BMI composite.

2000). The CTE of our composite measured experimentally 'in house' was confirmed by the manufacturer.

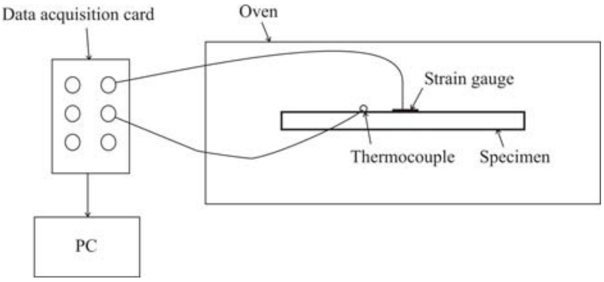

Fig 9 - Experimental set-up to measure strain and temperature.

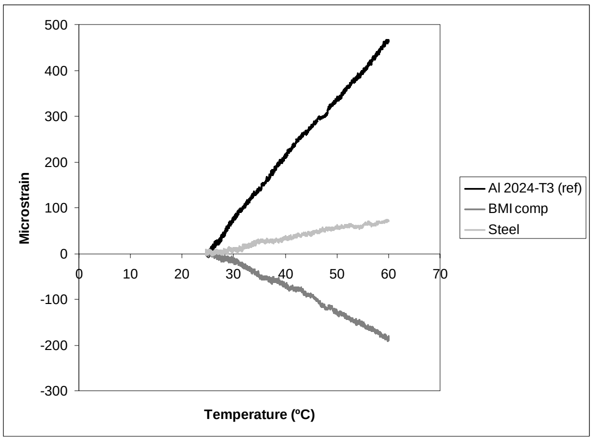

Fig 10 - CTE raw data results by a strain gauge technique.

6 - CONCLUSIONS

The mechanical properties of a carbon reinforced bismaleimide were measured from -55 to 200ºC using non conventional techniques. The main conclusion is that the through thickness strength is very low (one order of magnitude) in comparison with the longitudinal strength. This can cause problems, especially when the composite is loaded in the through thickness direction such as in adhesive joints.

REFERÊNCIAS

ANSI/ASTM D 790 – 71, Standard test methods for flexural properties of plastics and electrical insulating materials, 1971.

da Silva and Adams, Joint strength predictions for adhesive joints to be used over a wide temperature range, in preparation, 2005

Lord J.D., Strain gauge techniques for measuring thermal expansion. NPL Report CMMT(MN)012, 1997.

Adams R. D. and Gaitonde J. M., Low-temperature flexural dynamic measurements on PEEK, HTA and some of their carbon fibre composites Composites Science and Technology **47**, 271-287, 1993.

Rojstaczer S., Cohn D. and Marom G., Thermal expansion of Kevlar fibres and composites. J. Mater. Sci. **4**, 1223-1236, 1985.

Curtis P. T., Crag test methods for the measurement of the engineering properties of fibre reinforced plastics. *Royal Aerospace Establishment TR 88012*, 1988.

Hull D. and Clyne T. W. An introduction to composite materials, 2nd ed. Cambridge: Cambridge University Press, 1996.

Dieter G. E. ed., ASM handbook Vol. 20 Materials selection and design. Metals Park, Ohio: ASM International, 1997.

Ganesh V. K., Naik N. K., Thermal expansion coefficients of plane-wave fabric laminates, Composites Science and Technology 1994; **51**: 387-408.

Kim R. Y., Crasto A. S., Shoeppner G. A., Dimensional stability of composite in a space thermal environment, Composites Science and Technology 2000; **60**: 2601-2608.