MECHANICAL BEHAVIOR OF CF/POLYMER COMPOSITE LAMINATES UNDER CRYOGENIC ENVIRONMENT

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SUMMARY: The basic mechanical characteristics of carbon fiber reinforced plastics (CFRP) material systems are experimentally evaluated to discuss their applicability to the cryogenic propellant tanks of future reusable launch vehicles. The materials are based on different types of epoxy matrices, bismaleimide matrix and PEEK. The temperature dependent anisotropic elastic constants and thermal expansion coefficients are experimentally obtained and used in the analytical predictions of delamination propagation and matrix crack onset. Static tensile tests revealed that the matrix cracks tend to take place at drastically lower mechanical load under cryogenic environment, posing the possibility of fuel leakage through the chain of these matrix cracks. Interlaminar fracture toughness measurement showed that the toughness tend to increase at cryogenic condition. Numerical predictions of delaminations and matrix cracks based on the simple energy release rate calculations are shown to be in good agreement with the experimental results.

KEYWORDS: Cryogenic Environment, Liquid Propellant Tank, Mechanical Properties, Tensile Strength, Fracture Toughness, Delamination, Matrix Crack, Propellant Leakage.

INTRODUCTION

The wide use of composite materials is the major technical challenge for effectivelly reducing the structural weight of the planned future reusable launch vehicles. The cryogenic propellant tanks are the dominating structural components of the vehicle structure and thus the application of carbon fiber reinforced plastics (CFRP) to these components is one of the most promising technologies for realizing the aimed weight reduction. Investigation of the mechanical properties of CFRP under cryogenic temperature is essential in the considerations of the basic design concept of the vehicle. The goals of this study are firstly to investigate the temperature dependent mechanical properties of the epoxy-based, bismaleimide-based and PEEK-based CFRPs, and secondly to analyze the laminate behaviors under cryogenic environment. It is also the purpose of this study to look into the possible onset and accumulation of delaminations and transverse cracks that can affect the propellant permeability performance of the tanks.

MATERIALS AND EXPERIMENTAL SETUP

The material systems of CFRP used in the present investigation are based on 4 types of matrix polymers, 180 ^{o}C- and 120 ^{o}C-cured epoxy, bismaleimide and PEEK. They are purchased from 3 different suppliers. The material type identification using these matrix materials is given in Table 1.
The loading system utilizing 4-axes turret tensile system surrounded by the cryogenic chamber has been developed and installed to the Instron 4505 Testing Machine. This device facilitates up to 4 successive loadings of the specimens under single setup of the cryogenic environment. The tensile strains were measured by the clip-on type extensometer which is more reliable than the conventional strain gauges under cryogenic environment. The acoustic emission (AE) measurement was also utilized to detect the development of damages. This supplements the inability to directly observe the damages by optical means, as the whole loading system must be covered by the container throughout the experiment to realize the cryogenic environment. Real time observations of the specimen edges were made only under room temperature using the fiberscope optical observation system. Double cantilever beam (DCB) specimens were also prepared to measure the interlaminar fracture toughness, with the same loading system and chamber as for the tensile test being used. The delamination crack length was measured by attaching the crack gauges to one side of the specimen.

Table 1: Material types

	IM* CF/ 180 ^{o}C-cure epoxy	Standard CF/ 130 ^{o}C-cure epoxy	Standard CF/ Bismaleimide	IM CF/ PEEK
Supplier A	*Aa*	*Ab*	*Ad*	*Ae*
Supplier B	*Ba*	*Bb*	—	—
Supplier C	*Ca*	—	—	—

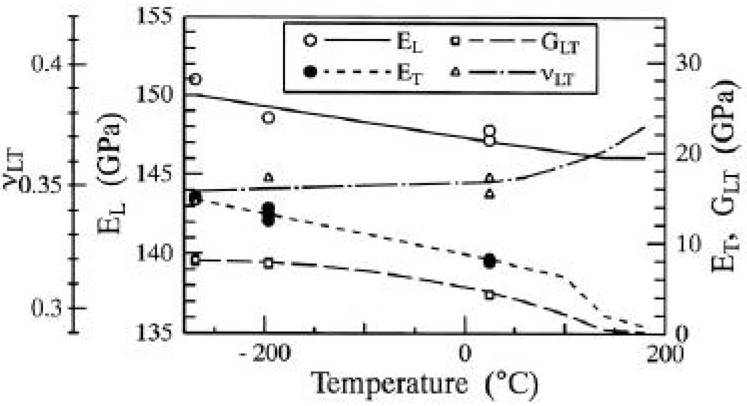
*IM : Intermediate Modulus

MECHANICAL PROPERTIES

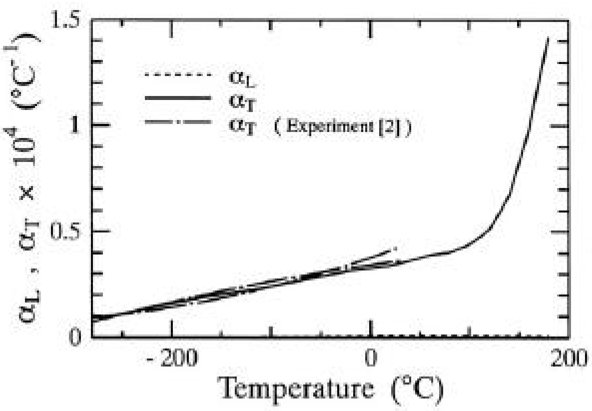
Elastic constants and thermal expansion coefficients

The existing data of cryogenic mechanical properties are often provided for specific discrete temperatures such as LN_2(-196^{o}C) or LHe(-269^{o}C) conditions [1]. In this respect, it is worthwhile to present the temperature dependent mechanical properties including thermal expansion coefficients and anisotropic elastic constants in the material principal directions as functions of temperature. The elastic constants of reference material type *Aa* which is the combination of intermediate modulus carbon fiber and 180 ^{o}C-cure toughened epoxy, are obtained by the tensile experiments conducted under discrete cryogenic temperatures and then extrapolated to the continuous functions of temperature based on the existing reference data. Unidirectional tensile specimens with fiber directions of 0^{o}, 45^{o} and 90^{o} were prepared and subjected to the static tensile tests. Thermal expansion coefficients were rearranged from the experimental results of reference [2] in which the same material type was employed.

Figure 1 shows the temperature dependent continuous variations of the material constants, together with the experimental results. In Fig.1(*a*), the experimental data points are shown by the symbols whereas the extrapolated values are shown by the corresponding curves. The principal directions are defined as fiber- (L) and transverse- (T) directions. The values of E_L and α_L are much less susceptible to temperature change than E_T, G_{LT}, G_{TT} and α_T, which can easily be predicted from the facts that the former properties are governed mainly by the fibers and that these fibers are merely affected by the changes in thermal condition. These data are used in the calculations of energy release rates and other parameters for predicting laminate performances including stiffness, strength and damage evaluation.



(*a*) Elastic constants



(*b*) Thermal expansion coefficients

Fig. 1: Temperature dependent material constants of CF/epoxy (material type *Aa*)

Tensile strength

Laminate behaviors under tensile loading have been experimentally investigated employing multiple material types shown in Table 1. The specimens used herein is fixed to $(45/0/-45/90)_{2S}$ quasi-isotropic, 16 ply, symmetric stacking configuration to simplify the comparison. The nominal specimen thickness and width were *2.2mm* and *15mm*, respectively. The tests were conducted under LN_2, LHe and room temperatures. The thermal effect on the static tensile strengths of all types of material systems is summarized in Fig.2. Reduction in tensile strengths under cryogenic temperature of up to nearly 80% of those under room temperature was observed. This may be attributed to the initiations of matrix cracks and delaminations at lower tensile loads, due to the development of higher thermal residual strains in the laminate under cryogenic temperature.

The comparison between the real time observation of the specimen edges and AE counts under room temperature were made. There was an obvious coincidence among the peaks of AE counts and the onset of matrix transverse cracks and delaminations. The delamination propagation could also be detected through the change in the apparent stiffness of the laminate. Typical comparison between the stress-strain curve and the AE counts for type *Aa* specimen is shown in Fig.3, where the stiffness change representing the delamination propagation can be observed. These observations are used to determine the onset of matrix cracks and delaminations under cryogenic environment where direct optical observations were prevented by the cryogenic setup. The resulting stresses corresponding to the onset of the matrix cracks and delaminations, together with the static strength, are shown in Fig.4 for various material types, both under room and LHe tempreratures. The results for material type Ca are not included due to the lack of measurement data. Generally, both the onset stresses for matrix cracks and delaminations tend to decrease when the temperature is decreased from room temperature to cryogenic temperature. Note that for CF/Bismaleimide (type *Ad*), the stress level at the onset of matrix cracks is zero under cryogenic environment, which means that the cracks developed right after mechanical tensile load picked up. It should also be noted that although the CF/PEEK (type *Ae*) specimen performed well under room temperature, the matrix cracks and delaminations tend to initiate at drastically lower load levels under cryogenic environment.

Interlaminar fracture toughness

In order to look into the detailed characteristics of delamination and matrix crack behaviors, it is essential to obtain the fracture toughness under cryogenic conditions. The DCB specimens made from material type *Aa* were prepared. The thickness and width of the specimen were *4.5 mm* and *12.7mm*, respectively. The interlaminar fracture toughness in terms of energy release rate under room and cryogenic temperatures are plotted in Fig.5. Each plot represents the average value over the full measured crack length range of each DCB specimen. It can be seen from the figure that the fracture toughness at cryogenic condition is much higher than that under room temperature, which can be attributed to the increase in strength of the matrix resin.

PREDICTIONS OF DELAMINATIONS AND MATRIX CRACKS

Though the final strength are of great interest when investigating the basic nature of the cryogenic effects on laminates, the damages induced in the laminates prior to global failure are of importance when designing the propellant tanks. The matrix cracks and delaminations can affect the component life of reusable vehicles, or the chains of these damages can possibly provide major leakage paths for the explosive propellant and thus increase the propellant permeability.

The energy release rates for free-edge delamination propagation and matrix transverse crack initiation are calculated based on the material properties of type *Aa* obtained above. The authors assume that similar tendency will be seen for other types of materials considered in this investigation and thus type *Aa* was chosen for the following investigations. The results

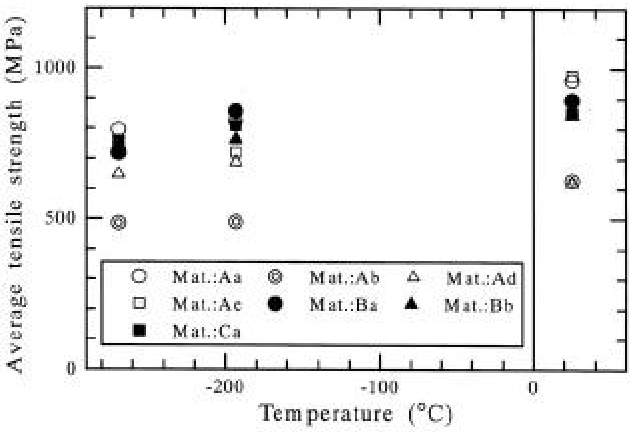
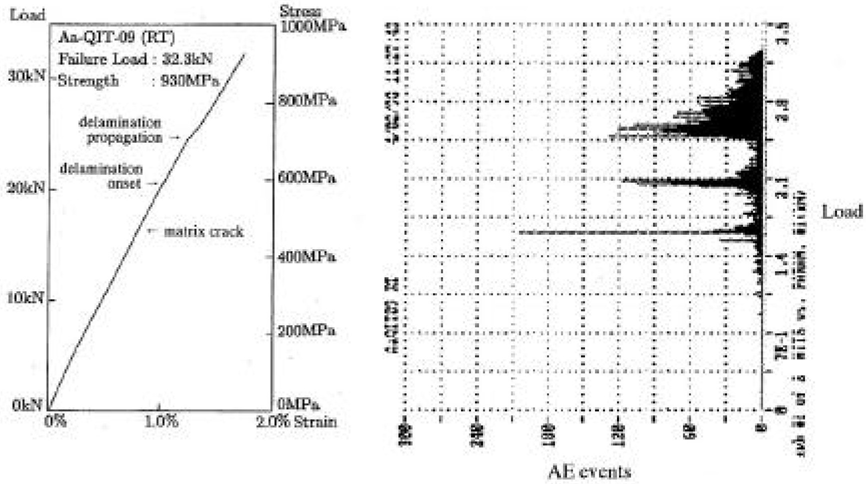
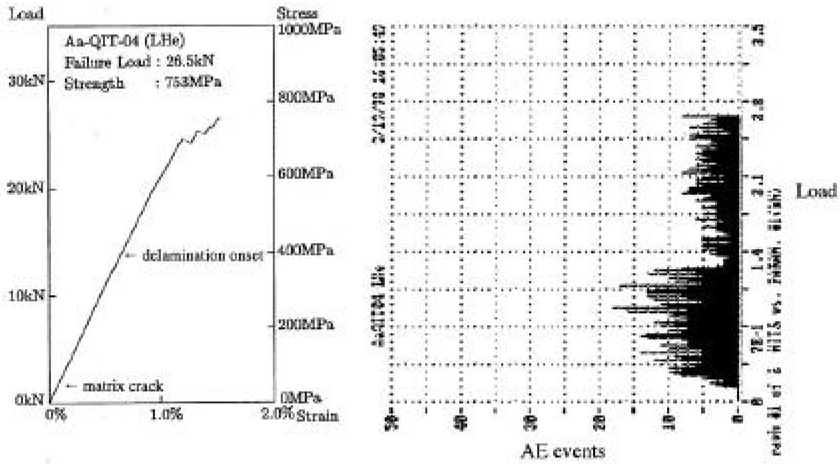


Fig. 2: Static tensile strength of quasi-isotropic laminates at different temperature

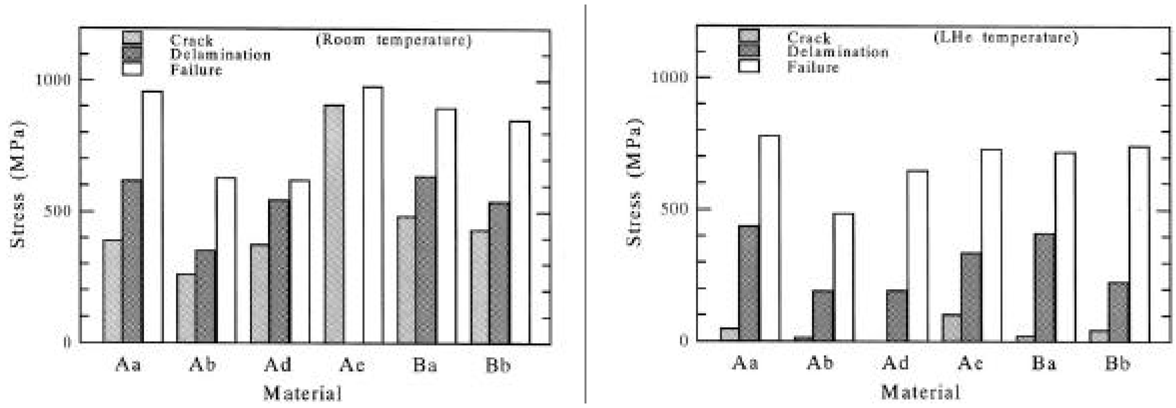


(*a*) Room temperature



(*b*) LHe temperature

Fig. 3: Tensile stress-strain curve and AE counts for CF/epoxy (material type *Aa*)



(*a*) Room temperature (*b*) LHe temperature

Fig. 4: Stresses at damage initiations and final failure (material type *Aa*, $(45/0/-45/90)_{2S}$)

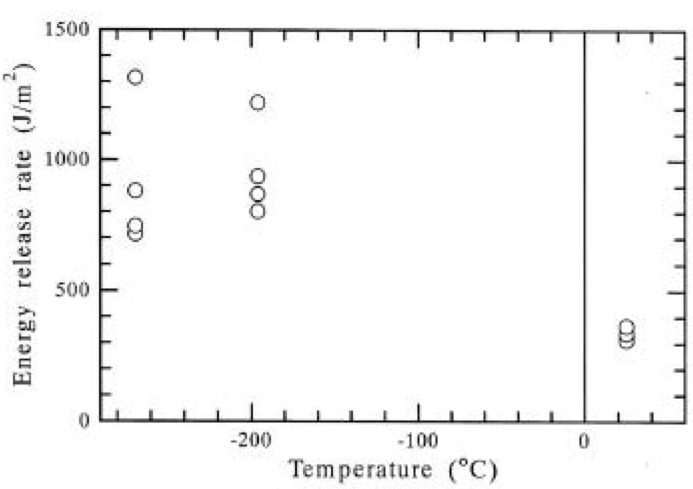


Fig.5: Interlaminar fracture toughness measured with DCB specimens (material type *Aa*)

show that the cryogenic conditions have little effect on the energy release rates for the delamination propagation, whereas the energy release rates for the matrix crack initiation greatly increase under the same cryogenic conditions. This implies that the matrix cracks tend to take place at comparably lower tensile load under cryogenic conditions. This is in good agreement with the experimental observations based on the optical survey and AE measurement discussed in the previous section.

The energy release rate calculation for free-edge delamination is based on the simplified method based on the free-edge superposition loads and J-integral proposed by Aoki and Kondo [3]. The calculated results based on this method are identical to those based on the stiffness method of O'Brien [4]. The thermal condition for free-edge delamination can be introduced as the thermal strain $\boldsymbol{e}_{Ti}$ as follows:

$$\varepsilon_{T_i} = \int_{T_0}^{T_0+\Delta T} \alpha_i(T)\,dT \qquad (i = L, T)$$

(1)

where T_0 is the temperature at no thermal stress or curing temperature, and *DT* is the temperature difference between T_0 and present or operating temperature. Appropriate elastic constants for given temperature are also used. In this study, T_0 is assumed to be 100 ^{o}C and the resulting temperature difference for room and LHe temperatures to be –80 and –370K.

For the energy release rate associated with the matrix cracks, the simple method based on shear-lag analysis presented by Park and McManus [5] was modified to incorporate both the mechanical and thermal loads simultaneously and applied to the present estimations. The final energy release rate (*G*) expression for the matrix crack is:

$$G = \frac{t_r t_c E_r E_c \Delta\varepsilon_{MT}^2}{2\xi(t_r E_r + t_c E_c)}$$

(2)

where E_r, E_c are average Young's moduli of intact layers and cracked layer, t_r, t_c are their thicknesses, *s* is the mechanical stress and *x* is the geometrical correction factor which is set to be *0.65* following the reference [5]. And De_t is the cumulative thermal strain difference between cracked and intact layers calculated as:

$$\Delta\varepsilon_{MT} = \frac{\sigma(t_r + t_c)}{t_r E_r} - \int_{T_0}^{T_0+\Delta T} \left(\alpha_c(T) - \alpha_r(T)\right) dT$$

(3)

The energy release rates associated with both the delamination propagation and transverse crack onset for given thermal conditions are summarized in Table 2 for material type *Aa*. Delaminations are assumed to take place at inner –45/90 interface of the $(45/0/\text{-}45/90)_{2S}$ laminate, and the matrix cracks at central 90_2 layers. Note that these values are for virtual delamination propagation or matrix crack onset assumed to take place at given temperatures and thus are the indications of susceptibility to the specified damage. The mechanical load corresponding to the tensile load that induces tensile strain of *5000m* at room temperature has been assumed in the calculations. The values in parentheses are those calculated on the assumption of constant material properties independent of temperature, which are fixed to the corresponding values at room temperature. This is to confirm the importance of utilizing the temperature-dependent material constants.

From Table 2, one can see that the energy release rates associated with delamination propagation is generally much smaller than those corresponding to matrix crack onset and the effect of the temperature dependence of material constants is trivial. On the other hand, the values associated with matrix crack are non-trivial, as the fracture toughness of the common toughened epoxy-based CFRP may spread over the range of 200-1500 J/m^2, depending on material, temperature, type of damage and other factors that may affect the toughness. Although the fracture toughness of CFRP tend to increase at cryogenic condition as shown above, the increase in energy release rate due to the higher thermal strain can override and may result in the matrix crack development at low temperatures. This also coincides with the experimental fact that the matrix crack onset takes place under very small tensile load at cryogenic environment.

The tensile stress at which the matrix crack is predicted to occur is plotted as functions of temperature in Fig. 6. The fracture toughness is assumed as a parameter in this figure. It can be seen that for relatively brittle material system, the matrix crack can take place under pure thermal loading with no mechanical loads.

The results strongly suggest that the design concept admitting the existence of matrix cracks should be adopted in order for the current CFRP to be reasonably applied to the cryogenic propellant tanks. The performance verification of laminates admitting the existence of matrix cracks must be conducted. Moreover, as discussed above, when the present material system is used for the cryogenic propellant tank, the chain of matrix cracks at each constituent layer, combined with possible delaminations, may provide a serious leakage path for the liquid propellant. Further experimental and numerical surveys are required to quantify the leakage in the light of the matrix cracks and delaminations.

Table 2: Energy release rates under assumed damage development

Virtual damage	Mechanical load (Inducing *5000m*at R.T.)	Room temperature (*DT=-80K*)	LHe temperature (*DT=-370K*)
Delamination	No	.4 (.2)*	20.1 (4.2)
	Yes	41.2 (38.4)	56.5 (60.8)
Matrix crack	No	15.2 (6.8)	260.7 (144.6)
	Yes	114.5 (88.5)	678.9 (354.7)

(Energy release rate in J/m^2)

*Figures in parentheses correspond to values calculated based on temperature-independent material constants.

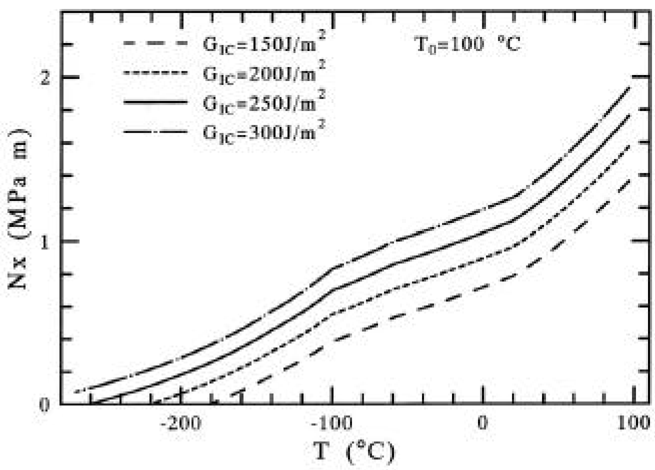


Fig.6: Predicted tensile stress at matrix crack onset as functions of temperature

CONCLUSIONS

The cryogenic performances of different types of CFRPs are experimentally evaluated to survey the basic applicability of the materials to the cryogenic propellant tanks. Temperature dependent material constants, tensile strength, interlaminar fracture toughness are experimentally obtained, together with detailed observations of matrix cracks and delaminations. The results indicate that the matrix cracks can possibly be one of the major critical issues when the current material system is considered for application to the cryogenic propellant tanks. The numerical predictions of the delaminations and matrix cracks are conducted to theoretically support the experimental consequences. The results suggest that possible application of reliable liner system must also be considered. At the same time the analytical scheme for evaluating the propellant leakage through matrix cracks existing in multiple layers in laminates must be developed to reduce the usage of liner materials for weight savings.

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