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# Mechanical Behavior of $Al_2O_3$ Dispersed CFRP Hybrid Composites at Room and Cryogenic Temperature

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$Al_2O_3$ particles were dispersed into carbon fiber reinforced epoxy composites to fabricate hybrid epoxy based composites. Interface behavior and mechanical properties of these hybrid composites were studied at room and liquid nitrogen temperature and the results were compared with those of carbon fiber reinforced composites to investigate their applicability at room and cryogenic temperature. Young's modulus in-perpendicular to fiber direction and interlaminar shear strength at room temperature and the thermal contraction down to cryogenic temperature were improved significantly by the addition of $Al_2O_3$ filler into the epoxy matrix. The effect of $Al_2O_3$ particle addition on mechanical properties were discussed.

***Key words:*** Carbon fiber, Hybrid composites, Cryogenic, Mechanical properties

## I. Introduction

Carbon fiber as a reinforcing agent have attracted much attention nowadays because of its high stiffness, high strength, low weight and good thermal properties. Carbon fiber reinforced composites(CFRP) mainly fiber in-parallel direction have been used in various fields especially in automobiles and marine industries at room temperature (RT) applications.[1] For low temperature applications it is used in aircraft body construction, communication satellite, space crafts, communication antennas, microwaves resonant cavaties, filters and telescope support structure.[2-3] However, the reduction of thermal contraction down to cryogenic temperature and strong interfacial bonding strength are considered as important properties for practical applications.

Fiber direction in the matrix plays an important role on the mechanical properties of CFRP. Mechanical properties of CFRP in-parallel to fiber direction is higher as the properties are mainly controlled by the fibers. However, the mechanical properties of CFRP in-perpendicular to fiber direction is inferior because the properties are mainly governed by the properties of the matrix used. Many researcher have investigated the mechanical properties of CFRP composites in-parallel to fiber direction, but only a few reports are available in-perpendicular to fiber direction. For practical applications, the properties in-perpendicular to fiber direction is also important for high performance structural design. Thus, it is important to study on the mechanical properties of FRP composites inperpendicular to fiber direction for practical applications at room and cryogenic temperature.

Interfacial properties or/and the interfacial strength also plays very important roles on the mechanical properties of CFRP composites. To produce CFRP composites with better mechanical properties, strong adhesion/bonding between the fiber and the matrix is required. Manocha[1] reported that interlaminar shear strength (ILSS) of CFRP composites can be improved by changing the matrix properties, suggesting that ILSS can be controlled by fracture mode of the matrix. However Nakao et al.[5-6] suggested that ILSS of CFRP composites depends on not only the properties of fiber and matrix but also the interfacial properties between the fiber and the matrix.

In this paper most attention has been paid to improve the mechanical properties of the matrix and the fiber-matrix adhesion strength. Thus for CFRP composites in-perpendicular to fiber direction and/or interface strength, attempt has been taken to improve the mechanical properties of the matrix and ILSS by adding $Al_2O_3$ filler particles into the epoxy matrix. The addition of $Al_2O_3$ filler into the matrix of CFRP are often designated as hybrid reinforced fiber composites and called hereafter carbon-fiber hybrid reinforced composites(CFHRP).

## II. Experimental Procedure

### 1. Materials

Carbon fiber reinforced composites (CFRP) and hybrid reinforced composites (CFHRP) were fabricated by filament winding method as described in our previous paper.[7] Poly Acrylic Nitrite (PAN) based carbon fiber from Toho Rayon Co., Ltd, Japan, as a main reinforcement, has been used as received. The epoxy matrix used in the fabrication process was N,N,N',N'- tetra-glycidyl-methoxy-diamine (TETRAD-X) produced by Mitsubishi Gas Chemical Co., Ltd. and 1,2-

cyclo-hexane-di-carboxylic-anhydride (HHPA) from Wako Jyunyaku Co. used as a curing agent. 10 vol% of $\gamma$-$Al_2O_3$ particles with an average particle size of 25 nm from Asahi Chemical Co. were used as a secondary reinforcing agent.

### *2. Composite testing*

Interlaminar shear strength (ILSS) of the composites was measured on an Instron Universal testing machine following the interlaminar shear compression (Guillotine) test method.[8] Under compression loading along the specimen length, the shear stress is distributed along the central plane section between the notches and causes fracture at a critical load. The crosshead speed was selected as 0.5 mm/min throughout the study. Test were carried out both at room (RT) and liq. $N_2$ (liquid nitrogen temperature). Fracture surfaces were examined by SEM observation to find out the effect of $Al_2O_3$ filler on the adhesion of fiber and the matrix at room and liq. $N_2$ temperature. Young's modulus in-perpendicular to fiber direction was estimated by resonance vibration method[9] at room temperature. Thermal contraction below to cryogenic temperature was measured using differential transformer thermal contraction measuring cell.[10] The thermal contraction was measured in-perpendicular to fiber direction.

## III. Results and Discussion

### *1. Young's modulus*

Fig 1 shows the Young's modulus in-perpendicular to fiber direction of CFRP and CFHRP composites as a function of fiber content. It was observed that Young's modulus of CFRP and CFHRP increased with increasing fiber content. Young's modulus of FRP in parallel to fiber unidirection depends on the modulus of fiber and the matrix of the composites which follow the rule of mixture in most of the cases. However, Young's modulus of the composites in-perpendicular to fiber direction depends mainly on the modulus of the matrix when the fiber volume fraction is constant. Thus the only way to improved the modulus of the composites in-perpendicular to fiber direction is to improved the modulus of the matrix. The rule of mixture is the easy and suitable method to calculate the modulus of the composites. Young's modulus of the composites in perpendicular to fiber direction could be calculated by the equation (1) as shown below.

$$1/E_c = V_f/E_{ft} + (1-V_f)/E'_m \qquad \text{eq.(1)}$$

$$E'_m = E_m/1-\upsilon^2 \qquad \text{eq.(2)}$$

Where $E_c$ represents the modulus of composites, $V_f$ is volume fraction of fiber, $E_m$ the Young's modulus of the matrix and $E_{ft}$ the Young's modulus of fibers in transverse direction (16 GPa), $\upsilon$ the Poisson's ratio of matrix. In this experiment Young's modulus of the matrix for CFRP was evaluated as 4.6 GPa and 6.86 GPa for CFHRP composites respectively.

It is evident from the Fig.1 that Young's modulus of the composites are in good agreement with the rule of mixture. Slight deviation of modulus from the experiment value is because of fiber misorientation and fibers overlapping. It is also revealed from the Fig.1 that Young's modulus of CFRP increased about 40-45% with addition of only 10 vol% $Al_2O_3$ filler into the matrix. Significant improvement of Young's modulus of CFRP composites by the dispersion of nano-sized $Al_2O_3$ filler particles into the matrix was suggested to be mainly contributed by improved Young's modulus of the matrix. Incorporation of nano-sized $Al_2O_3$ filler into the matrix were kept 10 vol% due to difficulty arise in fabrication processing over 10 vol% of $Al_2O_3$ filler dispersion.

### *2. Thermal contraction*

Thermal contraction of CFRP and CFHRP composites

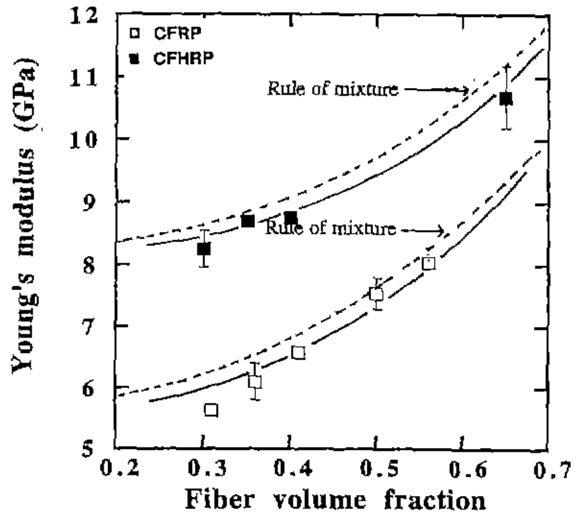

**Fig. 1.** Young's modulus as function of fiber content of CFRP and CFHRP at room temp.

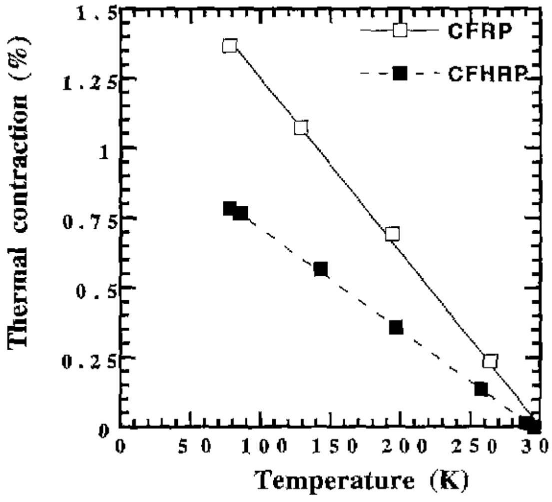

**Fig. 2.** Thermal contraction as a function of temperature of CFRP and CFHRP.

variation with temperature in perpendicular to fiber direction down to cryogenic temperature were measured and the results are shown in Fig. 2. It was observed that thermal contraction in-perpendicular to fiber direction measured down to liquid $N_2$ temperature for CFRP was 1.4%, whereas it was reduced to 0.80% by addition of 10 vol% $Al_2O_3$ filler into the matrix which indicates about 75% improvement by filler addition. Anisotropic properties of carbon fibers leads to a negative thermal expansion coefficient in fiber direction. However, when the thermal contraction of CFRP in-perpendicular to fiber direction is measured, fibers expands in-parallel direction and the fibers shrink in radial direction. Thermal contraction of epoxy down to liq. $N_2$ temperature was found to be approximately 0.97% and was smaller than that of CFRP in perpendicular direction. This is the evidence that the carbon fiber shrink in radial direction with cooling. Addition of 10 vol% of filler reduced the thermal contraction on cooling and thus improved the thermal contraction of CFRP composites and it is very important in practical applications. Dispersion of $Al_2O_3$ filler into the matrix decreases the mobility of epoxy network and increases the hardness of the composites, which is the main reason why the addition of $Al_2O_3$ filler improved the thermal contraction down to cryogenic temperature.

### *3. Interlaminar shear strength (ILSS)*

The strength or adhesion between fiber and matrix in CFRP composites can quantitatively be measured by interlaminar shear test. Fig.3 shows the variation of ILSS as a function of fiber content tested at room and liq. $N_2$ temperature. It was observed that both materials of CFRP and CFHRP carbon fiber content upto 0.36 volume fraction, showed an increasing tendency of ILSS with increasing fiber content. Moreover, CFHRP composites showed the significant improvement of ILSS as compared to CFRP at room temperature. These significant improvement was suggested for the improvement of matrix strength, strong interfacial bonding between the fiber and the matrix due to thermal residual stress during fabrication, and mechanical interlocking during fracture which absorbs high fracture energy in the presence of $Al_2O_3$ filler in the epoxy matrix.

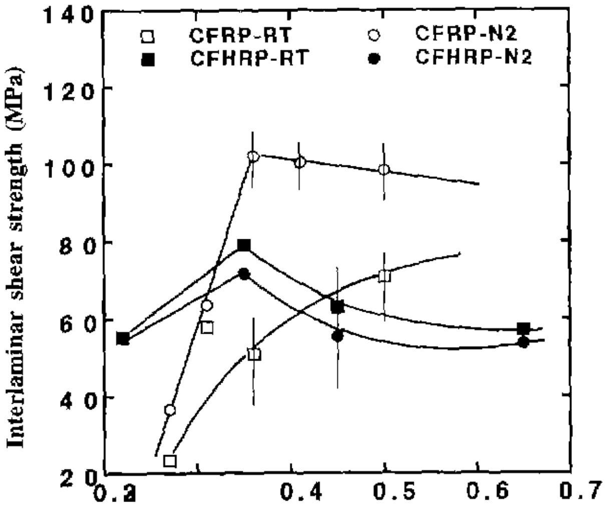

**Fig. 3.** ILSS of CFRP and CFHRP at RT and lid.$N_2$ as a function of fiber content.

**Fig. 4.** SEM micrographs of (0.36 vol.fraction of fiber) CFRP and CFHRP at Room temperature after ILSS test.

According to Outwater[11] when external load is applied, the shear transfer between the fiber and the matrix is resisted until the shear stress of $\tau = \mu P$. Where $\mu$ represents the interfacial friction coefficient and P represents the interface pressure on the fiber. However, interfacial coefficient of friction ($\mu$) is greatly depend on the surface roughness of the fiber. Clean fiber surfaces was observed by SEM for CFRP composites in Fig. 4(a) after ILSS tested at RT. Whilst considerable amount of matrix residues on the fiber surfaces of CFHRP composites was observed in Fig. 4(b). The presence of epoxy adhesion on the fiber surfaces clearly indicates the strong interfacial adhesion of epoxy matrix on the fiber surface in CFHRP composites. Furthermore, epoxy adhesion on the fiber surface of CFHRP composites

**Fig. 5.** SEM micrographs of CFRP (0.36 vol.fraction of fiber) after ILSS test at room and liq. $N_2$ temperature.

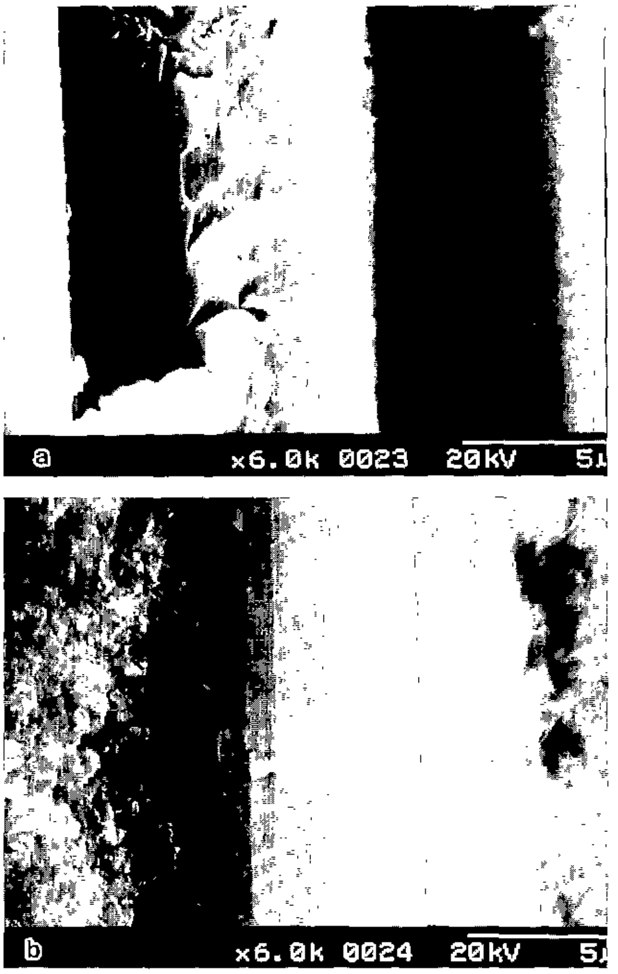

**Fig. 6.** SEM micrographs of (0.36 vol.fraction of fiber) CFRP and CFHRP after ILSS test at liq. $N_2$ temperature.

causes higher internal frictional coefficient and mechanical interlocking which causes higher ILSS value. It is believed that, for CFHRP composites $\tau$ is greater then that of $\mu P$ upto 0.36 volume fraction of fiber content but decreases later for higher volume fraction of fiber content as the interfacial surface area and the interface pressure (P) decreases and fiber-fiber interface area and fiber misalignment increases.

If there is no chemical bonding between the fiber and the matrix, adhesion is occurred due to Van der Waal's interactions[12] and change in thermal shrinkage of two different materials creates pressure at the fiber-matrix interface which attributes strong adhesion between fiber and the matrix. At liq. $N_2$ temperature CFRP composites showed the higher ILSS compare to RT results. It was observed that ILSS of CFRP showed higher value than that of CFHRP at liq. $N_2$ temperature. SEM micrographs of the fiber surfaces after ILSS test at liq. $N_2$ temperature can provide the valuable information. Fig. 5(a) and 5(b) show the micrographs of CFRP composites tested RT and liq. $N_2$ temperature respectively. Two distinguishable fiber surface was observed. Composites tested at RT shows a clean fiber surfaces which suggested weak adhesion between the fiber and the matrix. Crack was propagated through fiber-matrix interfaces. However when test were carried out at liq. $N_2$ matrices are strongly adhering on the fiber surfaces was observed clearly. When load was applied during ILSS test epoxy matrix are drawing out like fibrous materials. These suggested the strong interfacial adhesion between fiber and the matrix, higher interfacial pressure on the fiber surface caused by cooling and also the high strength of matrix itself due to compactness of epoxy network structure which promote the tailoring of ILSS. Cracks are shown to be propagating through the matrix and also fiber matrix interface. Mixed mode of fracture was suggested the one of the main reason for higher ILSS of CFRP down to cryogenic temperature.

However, CFHRP composites down to cryogenic temperature showed a lower ILSS value compared to CFRP composites. Fig. 6(a) and (b) represents the fracture surfaces of CFRP and CFHRP after ILSS test at liq. $N_2$ temperature. Fiber surface for CFHRP composites are shown clean without epoxy adhesion. At cryogenic temperature the interfacial strength between fiber and the matrix depends mainly on matrix strength and the thermal contraction of the matrix on the fiber. It was revealed that thermal contraction of CFHRP composites decreased by addition of $Al_2O_3$ filler into the matrix, thus the pressure exerted by the matrix decreased compared to CFRP which contribute lower ILSS of CFHRP at liq. $N_2$ temperature.

## IV. Conclusion

Young's modulus and thermal contraction in-perpendicular to fiber direction at room temperature could be improved with addition of 10 vol% $Al_2O_3$ filler into the matrix. ILSS of CFRP composites at down to cryogenic temperature was increased about 2-fold which was attributed to higher thermal contraction of matrix which causes strong interfacial adhesion between the fiber and the matrix and mixed mode of fracture. CFHRP composites showed the higher ILSS at RT compared to CFRP composites which is attributed to the increased interfacial adhesion and internal friction coefficient, thermal residual stresses and improvement of matrix strength caused by $Al_2O_3$ addition into the matrix.

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