

## 論文

### Mechanical Properties of Thermoplastic Matrix Composites

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열가소성수지 복합재료의 기계적 성질

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#### ABSTRACT

Superiority of CF/PEEK to CF/Epoxy composites is demonstrated in two aspects : tension-tension fatigue and CAI property of thick plates. Delamination arresting due to high fracture toughness provides such a superiority. In compressive strain of T-stiffeners, the present results surpass the existing results of CF/PEEK. Before main topics, a brief overview of the situation of composite technology in Japan and NAL is given.

#### 1. Introduction

The state-of-the-art of composite technology in Japan is presented first for the convenience of the readers. Composite activities and an outline of the Composite Structure Testing Facilities recently founded in NAL are also introduced briefly. Research program of thermoplastic composites is one of the highest priority activities in NAL.

Weight reduction is the most crucial point in design of aerospace structures. Current reduction ratios of about 20% to conventional metal structures by common carbon/epoxy composites, however, are not fully satisfactory considering very high stre-

ngths of recent carbon fibers. One reason of the less reduction ratio could be ascribed to poor fracture toughness of carbon/epoxy systems. A low allowable strain for a CAI(Compression After Impact) strength[1] is one typical example of the criticality caused by the poor toughness. Therefore, a strong demand for tough composites is now arising in the aerospace fields. Carbon/thermoplastic composites, particularly carbon/PEEK, are capable candidate materials for solving such a problem.

A practical research program of 6 years is now undergoing in NAL where a fabrication of wing structure models of AS4/APC-2 and an evaluation of its performance are the final goals. In-house

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fabrication of coupon level was done in NAL[2] and fabrication of some advanced specimens were conducted by Fuji Heavy Industries Co. LTD (FHI). Some remarkable experimental results of this program are demonstrated in the present paper including high fatigue strengths of unnotched coupons of quasi-isotropic laminates, superior buckling properties of T stiffeners to the existing results[3], and a verification and understanding of the process of high CAI strengths of thick plate specimens.

## 2. Overview of Japanese Composite Technology and NAL Activities

A short overview of Japanese composite technology is given here first where an emphasis is placed on the aerospace field.

In the technology of carbon(CF)/epoxy composites, Japanese level is considered to be reaching the maturing phase. Tailplanes of purely domestic T-4 military Trainer shown in Fig. 1 are fabricated by CF/epoxy. The new FSX program is highly related to CF/epoxy wings. Some CF/epoxy components for commercial transports such as parts of MD11 are in the level of daily production. In the space application, a majority of structural components of Japanese satellites is now made of high modulus CF/epoxy.

CF/thermoplastic systems and their application are in the developmental phase in Japan. NAL is playing the key role in this aspect. The technical essence will be shown in this paper.

Technology in high temperature polymeric composites like CF/polyimide is also under development. National Space Development Agency of Japan(NASDA) is now preparing its ambitious program of space transportation system called HOPE (Fig. 2). For assuring large payload, CF/polyimide is considered to be the best candidate material of HOPE's primary structure. NAL is suppor-

ting NASDA in the basic research.

Some other high-temperature composites are also investigated intensely in Japan. Carbon/Carbon(C/C) technology is developed to some extent. Ceramic/Ceramic technology is still in early phase. Functionally gradient material is also under development.

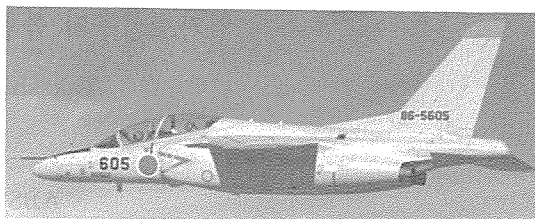


Fig. 1 T-4 Military Trainer

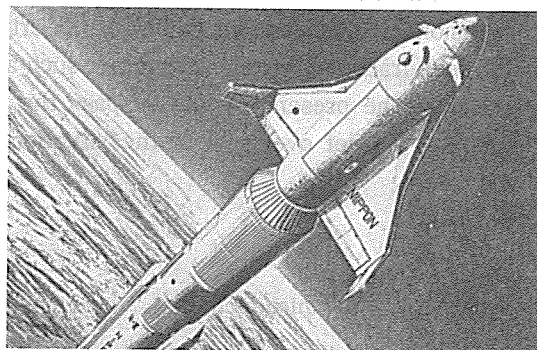


Fig. 2 An Artist's Impression of HOPE

As explained above, emphasized composite research programs in NAL are CF/TP, CF/PI and ceramic matrix composites. Moreover, NAL has been funded well recently in the field of composites in order to build its unique Composite Structure Testing Facilities. The total amount of money is 2.7 billion Yen for 5 years project. They consist of 3 major parts: the 1st part is Testing Machines for Specimen Level, the 2nd is Testing Machines for Structure Level, and the 3rd is Machines for Non-Destructive Evaluation. A typical NDE machine, Robotic Ultrasonic Scanning System, is

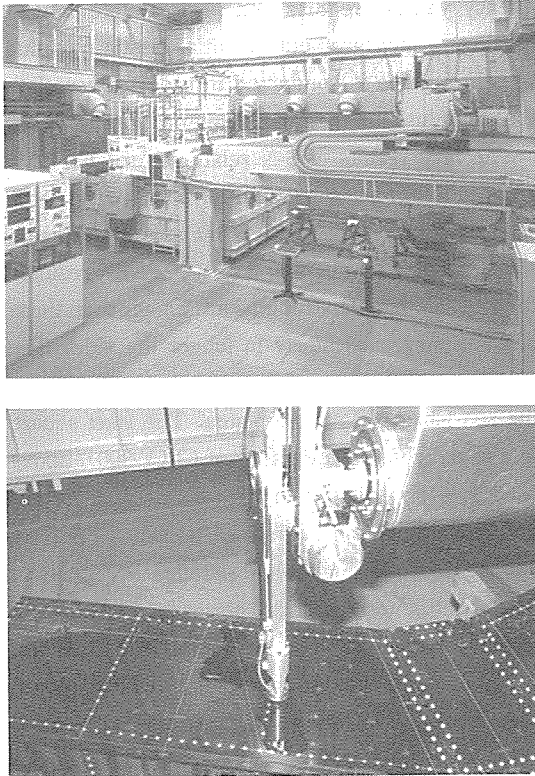


Fig. 3 Picture of Robotic Ultrasonic Scanning System

shown in Fig. 3 where above is a global view and below is a close-up of the probe. These brand-new facilities will be utilized pretty much for Japanese new era of composite technologies.

### 3. Background of Research of Thermoplastic Composites

Larger design allowables in stress or strain space can provide high ratio of weight reduction. A schematic diagram in the strain space quoted and modified from the original paper[4] is indicated in Fig. 4. The hatched zone in this figure corresponds to a typical CAI strain of carbon/epoxy in the 1-direction. Such a limit violates a hexagonal product set of the simple ply level allowable strain spaces

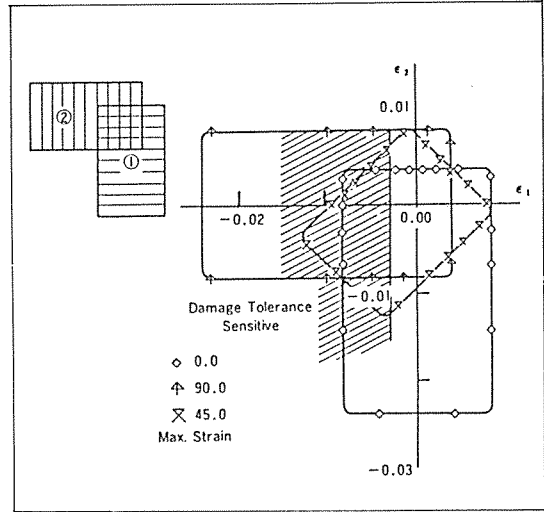


Fig. 4 Design Allowable Strain in Quasi-Isotropic Laminate.

described by rectangles. A crippling strain level in the spar of the carbon/epoxy horizontal tail model measured at the static strength test[5] is also quite close to this CAI strain. Some possible limits beyond ply level might be expected in the tensile side due to a free-edge delamination or a failure around a fastener. Thus, the size of the design allowable space tends to shrink if we consider some higher level criticals.

A detailed comparison of such off-ply level limits in both tensile and compressive properties between CF/PEEK and CF/epoxy composites is the objective of the present paper. Investigation of the strength of larger structures remains to be done for the work of near future.

### 4. Experiments in Tension-Tension Fatigue

As the first off-ply level property, a tension-tension fatigue strength for unnotched quasi-isotropic laminates is investigated here. Dimensions and 2 sorts of stacking sequence of the specimen are shown in Fig. 5. The same notation of the stacking

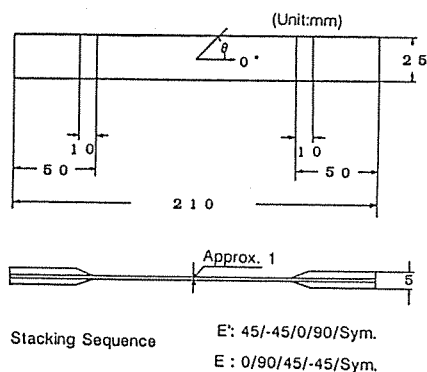


Fig. 5 Dimension of Specimens for Fatigue

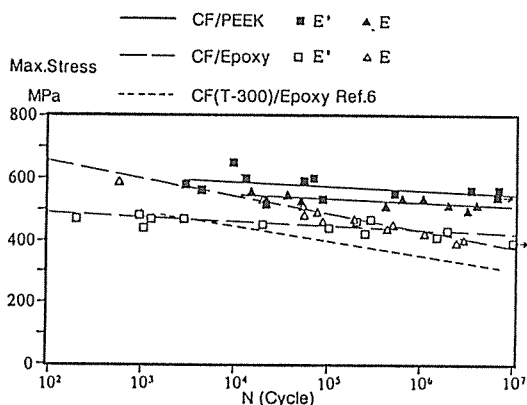


Fig. 6 Comparison of S-N Relations between CF/PEEK and CF/Epoxy

sequence as reference[6] is employed. The materials of interest and reference are AS4/APC-2 and T-400/3631, respectively. T-400 fiber was chosen as an equivalent to AS4. The in-house source plates of CF/PEEK are used and the tabs are bounded by FHI Inc. with an oxidizing surface treatment. The sectional shape of the tabs is a trapezoid. The specimens are loaded in tension by a servo-hydraulic machine of Shimadzu EHF 10 in a load control mode with a sinusoidal shape of a constant amplitude of the stress ratio of  $R=0.1$ . Employed fre-

quency values are 8Hz and 5Hz. Before fatigue loading, a measurement of initial elastic modulus by strain gages is conducted in order to correlate the modulus and strength later on.

The final results of S-N relations for CF/PEEK and CF/Epoxy are shown in Fig. 6 where the vertical axis corresponds to the peak stress. Although some scatter could be found for CF/PEEK results, their tensile fatigue strengths are higher than those of CF/Epoxy particularly in longer life side. The scatter is probably resulting from the quality of the self-fabricated source plates. The least square line of the reference data[6] of CF(T-300)/Epoxy (3601) is also plotted in Fig. 6. Because of the difference of carbon fibers, the present CF/Epoxy data show a little higher and parallel S-N line than that of reference[6]. It is, however, obvious that the inclinations of S-N lines for CF/PEEK and CF/Epoxy are quite different. High fracture toughness of CF/PEEK seems to cause this difference.

In order to examine such a presumption, delamination propagation is monitored by three types of method of non-destructive evaluation(NDE) during fatigue loading at an appropriate interval: The first method is a ultrasonic C-scan testing by a pulse echo technique utilizing Cannon M-500 system. Because the ultrasonic testing is dependent upon the setting of the pulse, and thus considered to be a relative measure in nature, a comparison to results of some other NDE techniques is inevitable. This point is the reason why two other methods are employed. The second method is a soft X-ray radiography by Softex SV-100AW system with a penetrant of DYB. The third method is a thermoelastic stress analysis by Spate 9000 analyzer. Developments of these NDE technique and their early evaluations are also a subsidiary purpose of the present research program.

Outputs of C-scan image for the stacking sequence of E' for the two materials are compared in

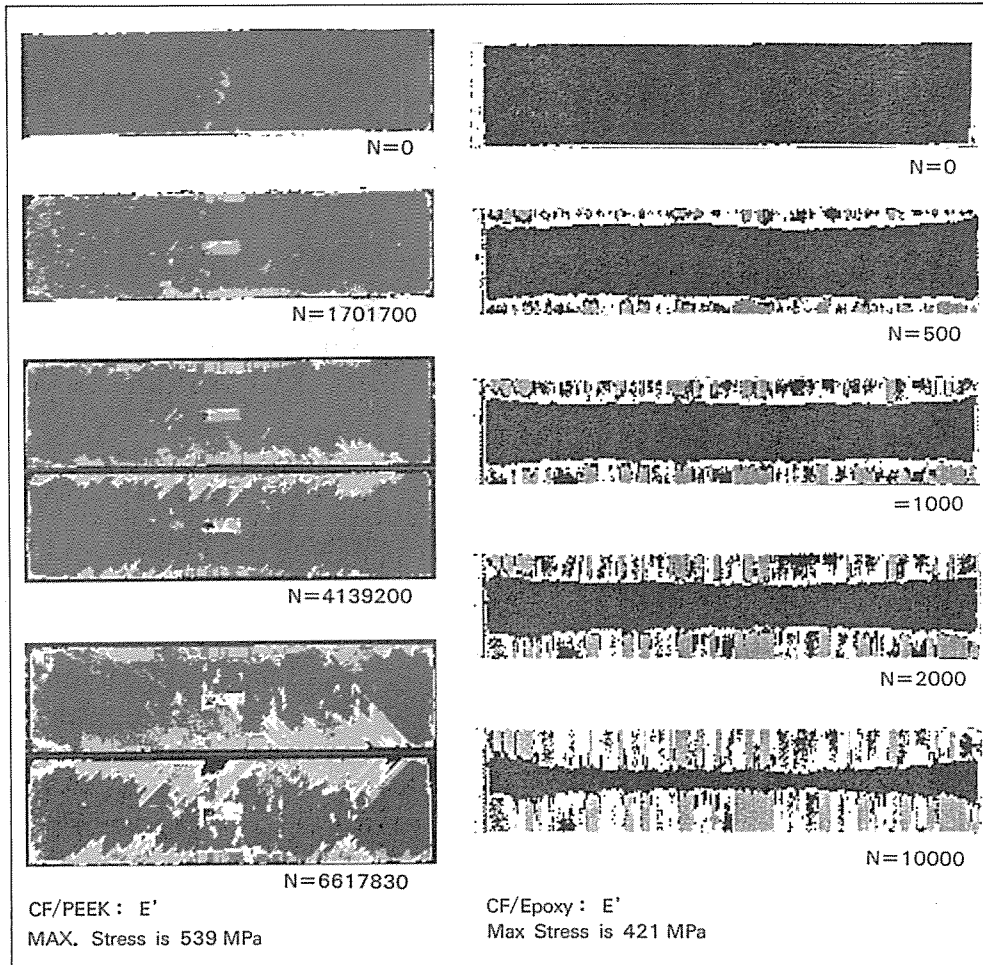


Fig. 7 Trace of Delamination Propagation by Ultrasonic C-Scanner

Fig. 7. In CF/PEEK for 539 MPa testing, central delamination at 0/90 interface is almost arrested in 3 or 4mm in depth. Next, delamination of Mode II and III mixture induced by the transverse cracks in surface 45 layers propagates slowly after a few millions at 45/-45 interface. In CF/Epoxy, the central delamination propagates very quickly even for lower peak stress of 421 MPa. Such an early delamination provides the lower strengths of E' in shorter life region indicated by open squares in Fig. 6.

For the stacking sequence E, the delamination behavior at 90/45 interface is quite different. It starts after a few million cycles in CF/PEEK and propagates rather fast. In Fig. 8, detected images by the 3 devices of NDE are shown comparatively for a 516 MPa case of CF/PEEK. Correlation between the images are very favorable. A rapid growth of the upper right corner leads to the final failure of the specimen. The stress concentration in the lower area of thermography outputs is resulted

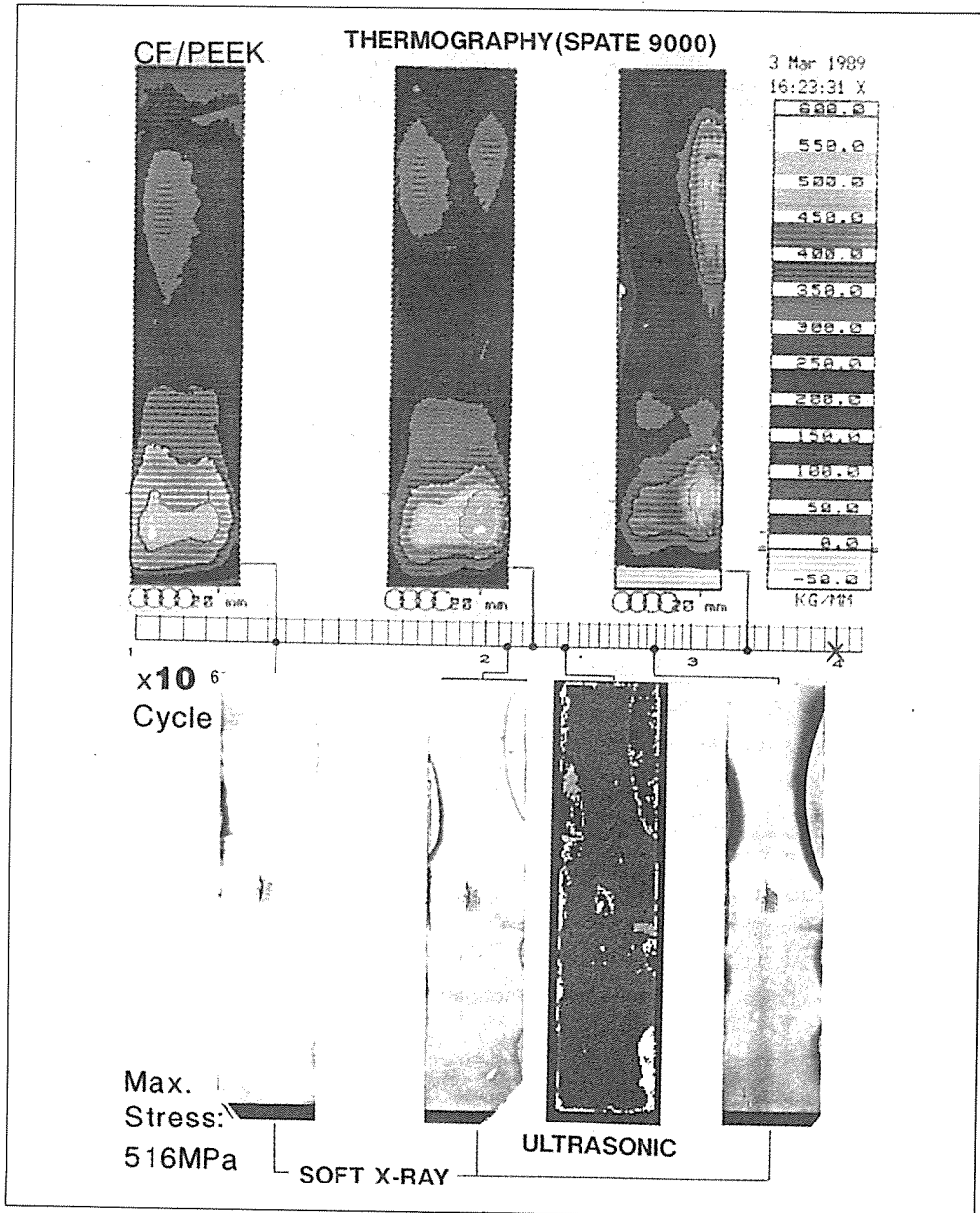


Fig. 8 Trace of Delamination by 3 NDE Methods.

from dense transverse crackings and longitudinal splittings. Even for sequence E, the delamination growth rate Fig. 8. Trace of Delamination by 3 NDE Methods is larger in CF/Epoxy. It can be summarized that high fatigue strengths of CF/

PEEK are brought by slow damage growth rate.

### 5. Compression Tests of T-Stiffeners

As one of a typical compressive design allowables

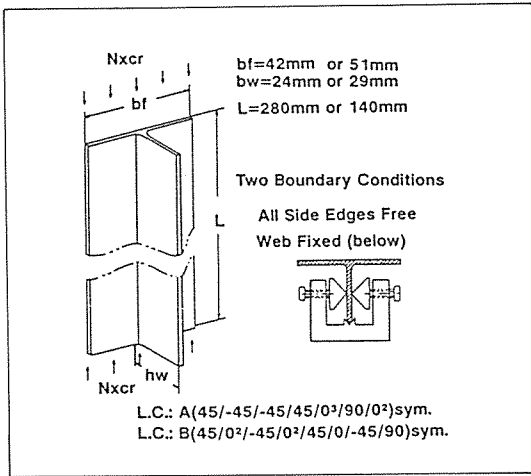


Fig. 9 T-Stiffener Specimen

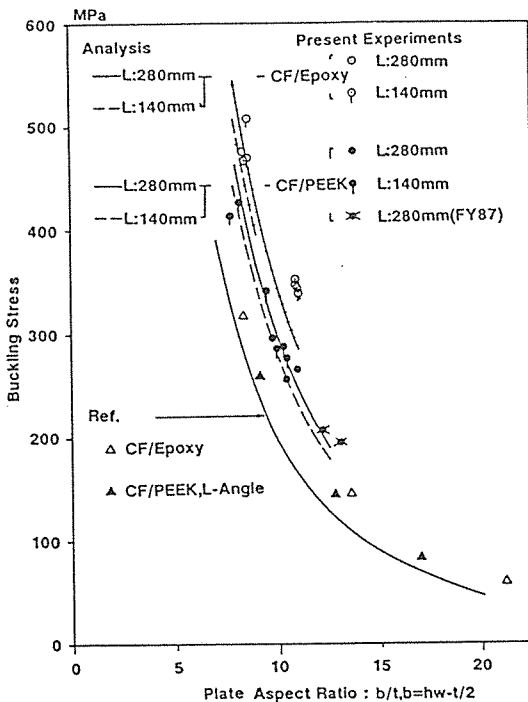


Fig. 10 Relationships between Buckling Stress and  $b_w/t$

for wing structure, postbuckling compressive strengths are chosen here. Because of the difficulties

of fabrication of T-stiffeners, it took several years to obtain the specimens of good qualities. The shape, dimensions and stacking sequence are indicated in Fig. 9. In order to examine the effect of the bending-torsion coupling,  $D_{16}$  and  $D_{26}$ , two types of the sequence are employed for CF/PEEK. It should be noted that the present sequence A which gives the better results is different from the sequence of reference (3) for L-stiffeners,  $(45/-45/0^4/45/-45/0^2)_{sym}$ . Two types of boundary conditions are also used here. However, the results for the fixed web condition will not be discussed in this paper. In CF/Epoxy (T-300/3601) test pieces, two values of flange width,  $b_f$ , are selected in order to examine the effect of width to thickness ratio,  $b/t$ .

The experiments are conducted by Instron 1128 machine without any interruption of crosshead movement. Strain data by 13 gages, lateral deflection of the web at the center, end shortening, and load data are recorded and stored at the sampling rate of 0.2 sec.

Discussions about buckling stresses are given first. Fig. 7 indicates a comparison between theoretical predictions and experimental results for buckling stresses for laminate A. Theoretical predictions are obtained by a simple Rayleigh-Ritz type method with the following form of deflection function[3] for a free-simply supported plate :

$$w(x, y) = \sum_{m=1}^3 \sum_{n=1}^3 \sum_{p=1}^3 \sin(m\pi x/a) \{ A_{mnp} \sinh(ny/b) + B_{mnp} \sin(p\pi y/b) \} \quad (1)$$

where  $a$  and  $b$  denotes the length and width of the plate, respectively. Using this function, the following description of the potential energy under an axial stress resultant of  $N_x$  can be calculated :

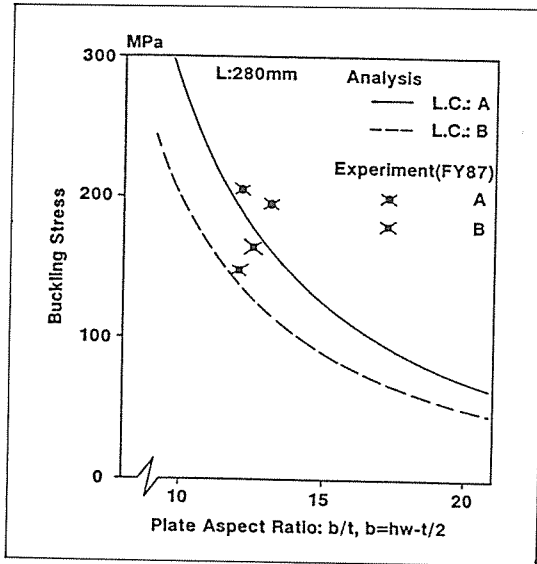


Fig. 11 Comparison of Buckling Stress in A and B Laminates

$$\Pi_p = \frac{1}{2} \begin{bmatrix} a & b \\ 0 & b \end{bmatrix} \{ D_{11}W_{,xx}^2 + 2D_{12}W_{,xx}W_{,yy} + D_{22}W_{,yy}^2 + 4D_{66}W_{,xy}^2 + 4D_{16}W_{,xx}W_{,yy} + N_x W_{,x}^2 \} dy dx \quad (2)$$

Then, by the stationary conditions of  $\Pi_p/\partial A_{mn}=0$  and  $\Pi_p/\partial B_{mp}=0$ , we can obtain an eigenvalue equation and buckling stress.

In order to obtain numerical results in Fig. 10, the following elastic moduli are employed for each material :

$$\begin{aligned} \text{CF/PEEK ; } E_L &= 120 \text{ GPa, } E_T = 10.4 \text{ GPa,} \\ G_{LT} &= 4.62 \text{ GPa, } \nu_L = 0.36 \quad (3) \end{aligned}$$

$$\begin{aligned} \text{CF/Epoxy ; } E_L &= 137 \text{ GPa, } E_T = 12 \text{ GPa,} \\ G_{LT} &= 6.6 \text{ GPa, } \nu_L = 0.34 \quad (4) \end{aligned}$$

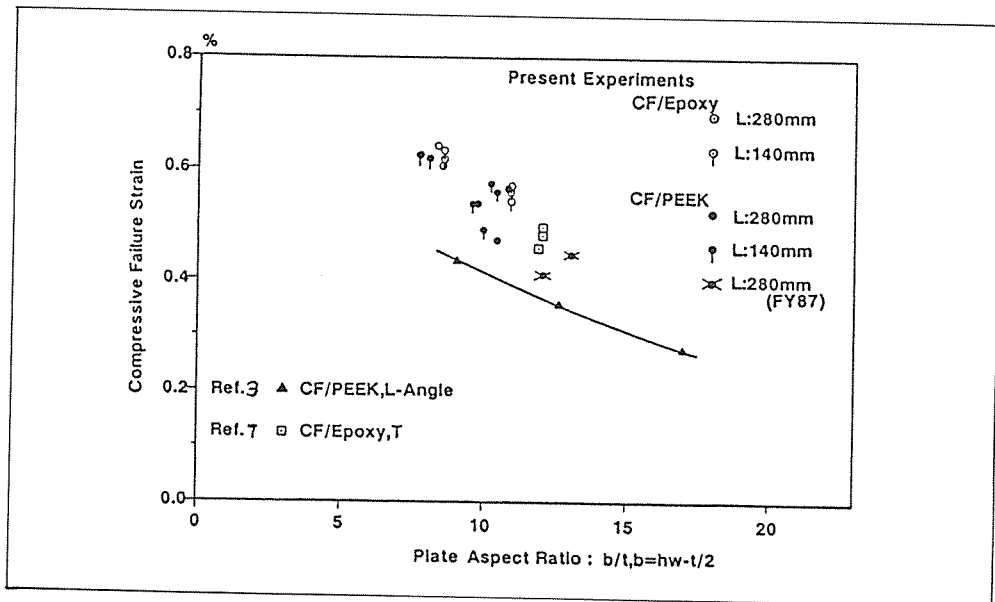


Fig. 12 Compressive Failure Strain vs.  $b_w/t$  Relationships in Various Stiffeners

For the re-calculation of the indicated curve for reference (3) for the same AS4/APC-2 system, the following moduli in that paper is adopted :

$$\begin{aligned} \text{CF/PEEK ; } E_L &= 110 \text{ GPa, } E_T = 8.2 \text{ GPa,} \\ G_{LT} &= 5.5 \text{ GPa, } \nu_L = 0.34 \quad (5) \end{aligned}$$



The values of CF/PEEK of eq.(3) is obtained in reference(2). High  $V_f$  in CF/Epoxy of 67% leads to larger value of moduli than those of CF/PEEK. The abscissa of Fig. 10 is a ratio of width to thickness,  $b_w/t$ , where  $b_w$  is defined by  $h_w - t/2$ . This definition implies that the center of rotation is considered to be the center of the intersectional area of the web and flanges. In the case of CF/PEEK, a variation of  $b_w/t$  is caused unintentionally, i.e., by a fluctuation of the thickness. Fig. 7 shows that the above simple prediction can provide a fair correlation to the experimental data. In the small  $b_w/t$  region, experimental values tend to be lower because of the effect of transverse shear. The reason why the present results are much better than those of reference (3) can be attributed to the sequence of the present laminate A where all  $\pm 45$  plies are placed to the outer regions. In the case of CF/Epoxy, this type of sequence leads to an early delamination onset and is not appropriate for actual structure, whereas it could be applicable to the structure in CF/PEEK due to high delamination resistance.

An effect of the stacking sequence upon buckling stress is more clearly shown in Fig. 11 where a comparison between the present A and B laminates for CF/PEEK is given. Filled circles with x are identical plots to those in Fig. 11. Theoretical values for  $b_w/t=12$  are obtained as follows by moduli of eq.(3) :

Laminate A : 207 MPa, b : 139 MPa ..... (6)

Averaged experimental values of two specimens are as follows :

Laminate A : 210 MPa ( $b_w/t=12.6$ )

Laminate B : 152 MPa ( $b_w/t=12.2$ ) ..... (7)

Thus, the correlation is favorable. Such difference is brought by the bending-torsion coupling terms,  $D_{16}$  and  $D_{26}$ . The following values are calculated for  $t=2\text{mm}$  and by eq.(3).

$$A : D_{16} \cdot D_{26} = 0.196,$$

$$B : D_{16} = D_{26} = 2.84(\text{Nm}) \dots\dots\dots (8)$$

As a conclusion of T-stiffener buckling section, failure strain vs.  $b_w/t$  plots obtained by initial axial modulus and strengths are depicted in Fig. 12. Filled triangles and open squares are the results of references (3) and (7). Higher compressive failure strains are provided by better buckling properties, and hence, by the present stacking sequence of A. Delamination resistant CF/PEEK system facilitates an employment of such a stacking sequence.

## 6. CAI Tests of Thick Plates

The final off-ply level design limit discussed here is the strength of compression-after-impact(CAI) of thick plates. Performances in damage tolerance of both types of composites are best compared by the CAI test. A subsidiary objective here is a detailed experimental description of the failure process in CAI tests.

A method by NASA[8] is adopted where a quasi-isotropic stacking sequence of 48 plies :  $[(45/0/-$

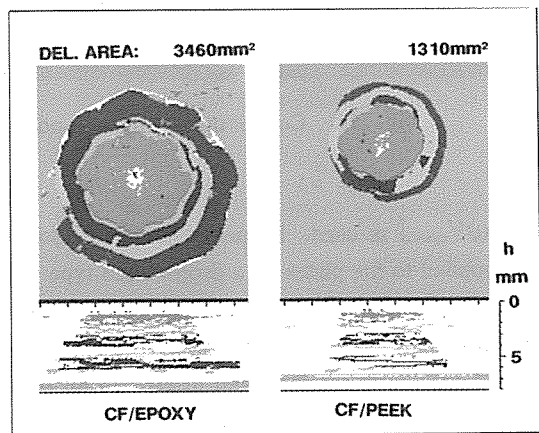


Fig. 13 Comparison of Delamination Area and Pattern in Both Composites

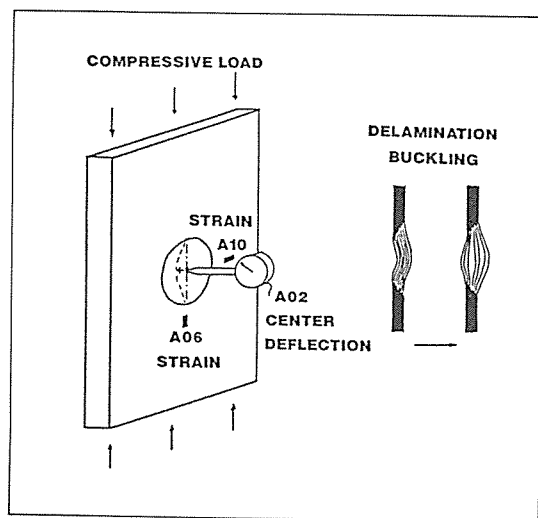


Fig. 14 Schematic View of CAI Test

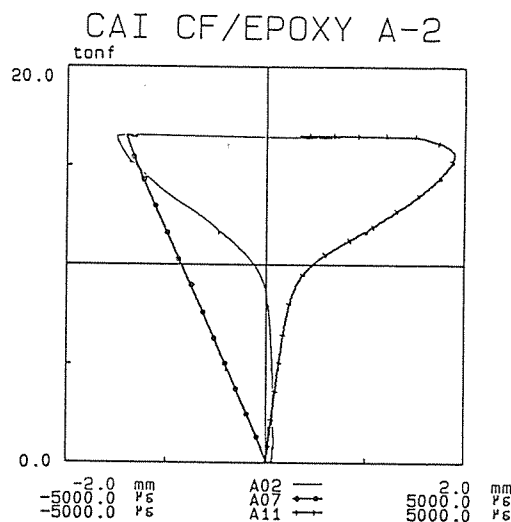


Fig. 15 Experimental Load vs. Strain and Deflection Curves for Impacted CF/Epoxy Plate

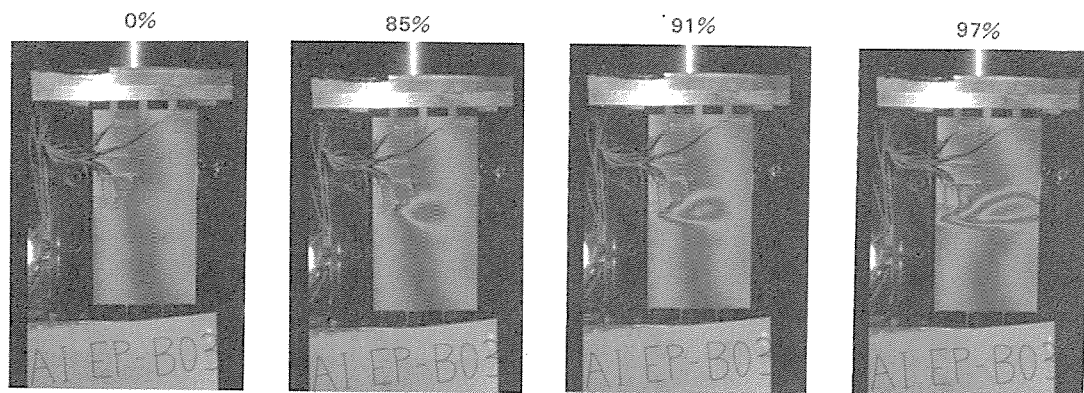


Fig. 16 Consecutive Moire-Topography Pictures for Impacted CF/Epoxy

45/90°<sub>sym.</sub>] is used. Impacts of about 27 Joule are applied to the plates by a drop-weight type impactor, Dynatup : GRC8250 system. Delamination patterns detected by the ultrasonic C-scanner explained previously are indicated in Fig. 13 for both materials. A larger area and a cone-like shape of delamination for CF/Epoxy is picked up in Fig. 13. In this section, AS/410 system of Mitsubishi Rayon Co. is selected as the CF/Epoxy material.

It is found that an area is smaller and a shape is more cylindrical for CF/PEEK.

Compression tests are conducted by Instron 1128 machine again for the plates after or before impacted. The end and edge fixtures are designed after reference (8). The loaded and supported ends are almost clamped and the side edges are almost simply supported. A schematic view of the positions of the deflection pickup and the selected strain cha-

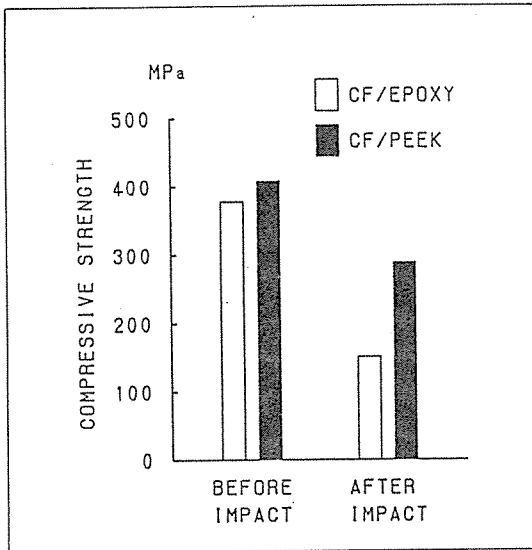


Fig. 17 Comparison in CAI Strengths of both Materials

nnels is given in Fig. 14 where an illustration of the final phase of the delamination buckling is also shown.

Fig. 12 depicts the behavior of these deflection and strain data for one of CF/Epoxy specimens after impacted. It is clearly shown in Fig. 15 that the center deflection measured from the side of the impact point (data : A02) is being reversed just before the final failure. This phenomenon is quasi-static and completely repetitive in impacted CF/Epoxy specimens. Neither in impacted CF/PEEK specimen, however, such a quasi-static deflection reverse occurs, nor in unimpacted specimens of both materials.

Consecutive moire-topography pictures taken from the other side of the impact point for CF/Epoxy is given in Fig. 16. Fringes indicate isovalue contour lines of the buckling deflection. Central fringe in a low load is intentionally given for a clear indication of an initial imperfection related to the delamination by impact. The numbers shown over Fig. 16 indicate percentages to the final load. This

figure describes the last stage of the CAI test where the buckling deformation induces a propagation of the delamination. The finally penetrated delamination seriously degrades a load-bearing capacity of the plate. Although an indication is omitted here, corresponding pictures for CF/PEEK clarify that the delamination penetration is well arrested just by the instance of final catastrophic failure that occurs at much higher stress.

For the conclusion of this section, CAI strengths are depicted in Fig. 17 as well as the strengths before impact. It is clarified that CAI strength of CF/PEEK is almost twice of that of CF/Epoxy of a conventional type.

## 7. Conclusions

Fatigue strengths of CF/PEEK are much higher than the corresponding CF/Epoxy laminates. Low rate of delamination growth cause such high strengths. Compressive failure strains of T-stiffeners of CF/PEEK exceed the existing value of reference (3). Concentrated  $\pm 45$  layers on the surface provide higher compressive strain. CF/PEEK system allows an employment of such a sequence. CAI strain of CF/PEEK obtained for thick plates is almost twice of that of CF/Epoxy. An arrest of delamination growth perpendicular to the loading axis gives the higher CAI strain of CF/PEEK.

## Acknowledgements

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