THE EFFECT OF INTERLAYER MODIFICATIONS ON TENSILE PROPERTIES OF HIGH STRENGTH PAN-BASED AND HIGH MODULUS PITCH-BASED HYBRID CARBON FIBER REINFORCED EPOXY MATRIX COMPOSITE

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Abstract

The tensile properties of the high strength PAN-based and high modulus pitch-based hybrid carbon fiber reinforced epoxy matrix composites with several types of interlayer modifications were investigated. For the Epoxy (EP), polyurethane (PU), acrylic (PMMA) films interlayer modified hybrid CFRP specimens, the tensile stress-strain curves of the hybrid CFRP specimen shows a complicated shape (jagged trace). The tensile responses of EP and PMMA films interlayer modified hybrid CFRP specimens were almost similar to those of unmodified hybrid CFRP specimen, although the holding stress of EP film interlayer modified hybrid CFRP was quite large. However, for PU and polyvinyil butyral (PVB) films interlayer modified hybrid CFRP specimens, the tensile properties were different from those of unmodified hybrid CFRP specimen.

1 Introduction

Fiber reinforced polymer matrix composites have become a dominant material in the aerospace, automotive and sporting goods industries [1]. Some of these composites are useful, however, only in highly specialized situations where limitations such as brittle fracture behavior are considered. By mixing two or more types of fiber in a common matrix to form a hybrid composite it may be possible to create a material possessing the combined advantages of the individual composite [2].

Polyacrylonitrile (PAN)- and pitch-based carbon fibers are widely used as a reinforcement in carbon fiber reinforced polymer matrix composites (CFRPs) because of their high specific strength and modulus [3-6]. Today, a number of high strength (more than 5 GPa) PAN-based and high modulus (more than 800 GPa) pitch-based carbon fibers have been commercially available and it is possible to create the high performance hybrid CFRPs. However, the mechanical properties of these hybrid CFRPs have not yet been well documented.

Recently, the tensile properties of high strength PAN-based (IM600) and high modulus pitchbased (K13D) hybrid CFRP were characterized by Naito et al [7]. As the results, the tensile stress-strain curve of the hybrid CFRP shows a complicated shape (jagged trace). By the high modulus pitch-based carbon fiber, the hybrid CFRP shows the high modulus in the initial stage of loading. Subsequently, when the high modulus carbon fiber begin to fail, the high strength fiber would hold the load (strength) and the material continues to endure high load without instantaneous failure. The observed stress after the initial fracture was improved by the enhancing the delamination energy.

In the present work, the high strength PAN-based (IM600) and high modulus pitch-based (K13D) hybrid CFRPs with several types of interlayer modifications were fabricated due to control the delamination phenomena. The tensile tests of these hybrid CFRPs were performed and to evaluate the potential of them.

2 Materials and testing methods

2.1 Materials

CFRP laminates were produced using a unidirectional CFRP prepreg material QC133-149A (fiber: IM600, resin: 133) and HYEJ16M95DHX1 (fiber: K13D, resin: HX1). IM600 and K13D carbon fibers were a high strength PAN-based and a high modulus pitch-based carbon fibers, respectively. IM600 (QC133-149A) prepreg was supplied from Toho Tenax Co., Ltd. and K13D (HYEJ16M95DHX1) prepreg was supplied from Mitsubishi Plastics, Inc. All sheets were manufactured using conventional prepreg technology. CFRP prepregs with a nominal thickness of 0.142 mm (QC133-149A, FAW (fiber area weight): 145 g/m², RC (resin content): 35 %) and 0.133 mm (HYEJ16M95DHX1, FAW: 160 g/m², RC: 33 %) were used.

Four different types of films (epoxy, polyurethane, acrylic and polyvinyil butyral resin) were used for the interlayer modifications. The epoxy adhesive film (EP; Redux312UL) was supplied from Hexcel Composites Ltd. The polyurethane adhesive film (PU; Thermoplastic Polyurethane Elastomer U-1490) was supplied from Kurabo Industries Ltd. The acrylic resin film (PMMA; Technolloy S001G) was supplied from Sumitomo Chemical Co., Ltd. and the polyvinyl butyral resin film (PVB; S-Lec B) was supplied from Sekisui Chemical Co., Ltd. The thicknesses of EP, PU, PMMA and PVB films are 83, 100, 100 and 380 µm.

2.2 Specimen preparation

The prepreg sheets and films were cut into the appropriate size. The sheets and films were placed on the vacuum molding board with plain woven fabric CFRP plate to reduce the thermal mismatch between the prepreg and the molding board. The hybrid CFRP laminates were made using hand lay-up and vacuum bagging technique (no bleeder). The laminates were pressed in 0.294 MPa and cured in 180 °C for 4 h (heating rate was 1 °C/min) by an autoclave (Ashida Mfg. Co., Ltd., ACA Series) in the laboratory. Fiber orientations of the unmodified interlayer modified and hybrid CFRP laminates were set to $[0_{(IM600)}/film/0_{(K13D)}/film/0_{(IM600)}/film/0_{(K13D)}/film/0_{(K13D)}/film/0_{(IM600)}/film/0_{(IM600)}]$ and $[0_{(IM600)}/0_{(K13D)}]_{2S}$. The fiber volume fractions of the hybrid CFRP laminates were 36.4 (EP, IM600: 19.1 vol%, K13D: 17.3 vol%, IM600 CFRP : K13D CFRP = 51.0 : 49.0), 34.0 (PU, IM600: 17.9 vol%, K13D: 16.1 vol%, IM600 CFRP : K13D CFRP = 51.0 : 49.0), 34.0 (PMMA, IM600: 17.9 vol%, K13D: 16.1 vol%, IM600 CFRP : K13D CFRP = 51.0 : 49.0), 16.3 (PVB, IM600: 8.6 vol%, K13D: 7.7 vol%, IM600 CFRP : K13D CFRP = 50.5 : 49.5) and 55.7 vol% (unmodified, IM600: 29.3 vol%, K13D: 26.4 vol%, IM600 CFRP : K13D CFRP = 51.6 : 48.4).

The hybrid CFRP laminates were cut into rectangular straight side tensile test specimens with dimensions of 200 mm in length (gauge length, L of 100 mm) and 10 mm in width. The fiber axis in the specimen was oriented in line with the length of the tensile test specimen (0° direction specimen). To remove the effect of stress concentrations caused by surface roughness from the edges, the edges of tensile test specimen were polished to remove the scratches. Thinner plain woven fabric glass fiber reinforced plastics (50 mm in length, 10mm in width and 1 mm in thickness) tapered tabs were affixed to the tensile test specimen to minimize damage from the grips on the tensile testing machine.

2.3 Tensile test

Tensile tests of the hybrid CFRPs were performed using a universal testing machine (Shimadzu, Autograph AG-series) with a load cell of 100 kN. The specimen was set up to the testing machine. The crosshead speed of 5.0 mm/min was applied. All tests were conducted under the laboratory environment at room temperature (at 23 ± 3 °C and 50 ± 5 % relative humidity). Strain gages were used to measure longitudinal strains. Five specimens were tested for the interlayer modified hybrid CFRPs (Twenty specimens were tested for the unmodified hybrid CFRP). The tensile modulus is calculated using a least square method for the straight line section of stress-strain curve.

3 Results and discussions

2.1 Materials

Figure 1 shows typical tensile stress-strain (σ - ε) curve for the interlayer modified and unmodified hybrid CFRP specimens. The stresses of the interlayer modified hybrid CFRP specimens were normalized to volume fraction of fiber, $V_f = 55.7$ % (unmodified hybrid CFRP specimen).



Figure 1. Tensile stress-strain curves for the interlayer modified and unmodified hybrid CFRP specimens.

For the EP, PU, PMMA films interlayer modified and unmodified hybrid CFRP specimens, the tensile stress-strain curves of the hybrid CFRP specimen shows a complicated shape (jagged trace). By the high modulus K13D CFRP layers, the hybrid CFRP specimen shows the high modulus in the initial stage of loading (this modulus was defined as tensile modulus, E_C). Subsequently, when the K13D CFRP layers begin to fail (this strength and strain were defined as initial fracture strength, σ_{Ci} , and initial failure strain, ε_{Ci}), the high strength IM600

CFRP layers would hold the load (strength) and the hybrid CFRP specimen continues to endure high load without instantaneous failure (this modulus was defined as secondary tensile modulus, E_{C}^{*}). The holding stress level is different among the EP, PU, PMMA films interlayer modified and unmodified hybrid CFRP specimens and these stresses were defined as holding stress, σ_{Ch} . Finally, the load reached its maximum and fracture of the hybrid CFRP specimen occurred (this strength and strain were defined as tensile strength, σ_{Cf} , and failure strain, ε_{Cf}). However, for the PVB interlayer modified hybrid CFRP specimens, the stress applied to the specimen was almost linearly proportional to the strain until failure (this modulus, strength and failure strain were defined as tensile modulus, E_C , tensile strength, σ_{Cf} , and failure strain, ε_{Cf}). The average tensile modulus (E_C), secondary tensile modulus (E_{C}^{*}), strength (σ_{Cf}), failure strain (ε_{Cf}), initial fracture strength (σ_{Ci}), initial failure strain (ε_{Cf}) and holding stress (σ_{Ch}) were shown in Table 1. For the mono (IM600 and K13D) composites, the tensile modulus (E_C), fracture strength (σ_{Cf}) and failure strain (ε_{Cf}) were also shown in this table.

	Tensile modulus E_C (GPa)	Initial fracture strength σ _{Ci} (GPa)	Initial failure strain \mathcal{E}_{Ci} (%)	Secondary tensile modulus E_{C}^{*} (GPa)	Tensile strength σ_{Cf} (GPa)	Failure strain ɛ _{Cf} (%)	Holding stress σ_{Ch} (GPa)
EP ineterlayer	334	1.365	0.411	70	1.546	1.769	1.195
modified *	±15	±0.069	±0.017	±6	±0.033	±0.023	±0.047
PU ineterlayer	277	1.069	0.407	70	1.533	1.637	1.038
modified *	±13	±0.039	±0.009	±4	±0.025	±0.017	±0.049
PMMA ineterlayer	320	1.238	0.393	73	1.539	1.684	0.755
modified *	±6	±0.028	±0.005	±6	±0.032	±0.016	±0.011
PVB ineterlayer	66				1.009	1.557	
modified *	± 1	-	-	-	±0.101	±0.061	-
Unmodified **	310	1.318	0.432	74	1.337	1.662	0.737
	±4	±0.034	±0.010	±3	± 0.068	±0.062	±0.022
IM600 **	158				2.718	1.675	
(QC133-149A)	± 3	-	-	-	±0.127	±0.059	-
K13D **	487				1.638	0.343	
(HYEJ16M95DHX1)	±7	-	-	-	±0.119	±0.031	-

* modulus, strength and stress were normalized to volume fraction of fiber, $V_f = 55.7$ % (unmodified hybrid CFRP specimen).

** from previous investigation [7].

Table 1. Tensile properties of the interlayer modified and unmodified hybrid CFRP specimens.

The tensile properties of EP and PMMA films interlayer modified hybrid CFRP specimens were almost similar to those of unmodified hybrid CFRP specimen, although the holding stress of EP film interlayer modified hybrid CFRP was quite large (σ_{Ch} of 1.195 GPa). However, for PU and PVB films interlayer modified hybrid CFRP specimens, the tensile properties were different from those of unmodified hybrid CFRP specimen. The tensile modulus and initial fracture strength of PU film interlayer modified hybrid CFRP specimen were slightly lower than those of unmodified hybrid CFRP specimen. For PVB film interlayer modified hybrid CFRP specimen, tensile stress-strain curve did not shows a jagged trace and tensile modulus, strength and failure strain were quite low.

The tensile modulus and secondary tensile modulus of the hybrid CFRP specimen were calculated using a simple rule of mixtures.

$$E_{C(Hybrid)} = E_{C(IM\,600)} V_{IM\,600} + E_{C(K13D)} V_{K13D}$$
 (for tensile modulus) (1)

$$E^*_{C(Hybrid)} = E_{C(IM\,600)} V_{IM\,600} \text{ (for secondary modulus)}$$
(2)

in which V_{IM600} and V_{K13D} are the volume fraction of the IM600 CFRP and K13D CFRP, respectively. The estimated tensile modulus, E_C and secondary tensile modulus, E_C^* of the hybrid CFRP specimen were 317 and 81 GPa, respectively. The experimental results were found to be in agreement with the rule of mixtures prediction. Similar results of hybrid composites were observed in some literatures [8-10].

The tensile strength and initial fracture strength of the hybrid CFRP specimen were also calculated using the rule of mixtures.

$$\sigma_{Ci(Hybrid)} = \sigma_{Cf(K13D)} V_{K13D} + \varepsilon_{Cf(K13D)} E_{C(IM \, 600)} V_{IM \, 600} \text{ (for initial fracture strength)}$$
(3)
$$\sigma_{Cf(Hybrid)} = \sigma_{Cf(IM \, 600)} V_{IM \, 600} \text{ (for tensile strength)}$$
(4)

The estimated tensile strength, σ_{Cf} and initial fracture strength, σ_{Ci} of the hybrid CFRP specimen were 1.402 and 1.067 GPa, respectively. The differences between the calculated and experimental results for tensile strength and initial fracture strength were 5 and 20 %. The experimental result of tensile strength was found to be in agreement with the rule of mixtures prediction. The final failure strain, ε_{Cf} as shown in Table 1, is also almost similar to that for the IM600 CFRP specimens. However, a large difference was observed in the initial fracture strength. The initial failure strain, ε_{Ci} as shown in Table 1, is also higher than that for the K13D CFRP specimens. Similar results of hybrid composites in which a phenomenon termed "hybrid effect" were also observed in some literatures [8-12].

4 Concluding remarks

The tensile properties of the high strength PAN-based (IM600) and high modulus pitch-based (K13D) hybrid carbon fiber reinforced epoxy matrix composites with Epoxy (EP), polyurethane (PU), acrylic (PMMA) and polyvinyil butyral (PVB) films interlayer modified hybrid CFRP specimens with fiber orientation of $[0_{(IM600)}/film/0_{(K13D)}/film/0_{(IM600)}/film/0_{(K13D)}/film/0_{(K13D)}/film/0_{(IM600)}/film/0_{(IM600)}]$ were examined. For the EP, PU, PMMA films interlayer modified and unmodified hybrid CFRP specimens, the tensile stress-strain curves of the hybrid CFRP specimen shows a complicated shape (jagged trace). By the high modulus carbon fiber (K13D), the hybrid CFRP specimens show the high modulus in the initial stage of loading. Subsequently, when the high modulus carbon fibers (K13D) begin to fail, the high strength carbon fibers (IM600) would hold the load (strength) and the CFRP specimens continue to endure high load without instantaneous failure. However, for PVB film interlayer modified hybrid CFRP specimen, tensile stress-strain curve did not shows a jagged trace and tensile modulus, strength and failure strain were quite low.

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