NANOFIBER INTERLEAVED COMPOSITES FOR AEROSPACE APPLICATIONS

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Abstract

This paper extended the authors previous work of demonstrating the concept of polymer nanofiber interleaving to enhance the toughness of fiber reinforced carbon/epoxy composite laminates. In this study, in-plane tension, compression, interlaminar shear, and Mode I fracture and fatigue onset life properties were studied. Results demonstrated that by adding 1% weight of nanofibers in the form of interlayer in-plane tension and compression properties (strength, modulus, and Poisson's ratio) of interleaved composites remained the same as base composites. Whereas polymer nanofiber interleaving increased interlaminar shear strength by 4%, fracture toughness by 150% and fatigue threshold energy release rate by 67%..

Keywords: *Fiber reinforced composite, polymer nanofiber, interleaving, and interlaminar toughness*

Introduction

Delamination [1] is a primary mode of failure in almost all fiber reinforced polymer composites. Solutions to control delamination, such as, matrix toughening [2], controlling stacking sequence [3], stitching [4, 5], braiding [5], edge capping [6] and ply termination [7] and adhesive interlayering [8, 9] are complex, expensive or not practical. Polymer nanofiber interleaving to enhance damping, impact toughness and fracture resistance was highlighted in Ref.10. This paper focuses on the impact of polymer nanofiber interleaving on in-plane properties, interlaminar shear and Mode I fracture onset life.

Electrospun Nylon-66 nanofiber [10] of about 1% wt of prepreg was placed between AS4/3501-6 prepreg layers and then the composite laminate was fabricated. In-plane tension and compression, interlaminar shear, and Mode I fracture and fatigue onset life properties were measured and compared with the base composite properties. Electrospinning process to produce Nylon-66 nanofibers of 100-200 nm diameter was explained in detail in Refs.10 and 11 and is not included here. Similar work was reported in literature with different polymer fibers, such as Polybenzimidazole [12], Polycarbonate [13], Nylon 6 [14], and epoxy EPO 1691-410 [15] with mixed success. The main problem in the compatibility of the nanofibers with the base epoxy and the amount of nanofibers.

Experimental

Materials

Aerospace grade AS4/3501-6 prepreg supplied by Hexcel Composites was selected for toughening with Nylon-66 nanofiber produced by electrospinning. Nylon-66 was supplied by Dupont and had molecular weight of 20,000 g/mol. Nylon-66 has extremely high elongation to fracture, high melting/softening temperature (250°C), and readily bonds with epoxy. Also, it survives autoclave process and the cure temperature of 177°C. Hence Nylon-66 was chosen for interleaving.

Nylon-66 dissolved in a mixture of formic acid and chloroform with a weight ratio of 75/25 was used to produce nanofibers using an in-house built electrospinning setup. Morpology of the nanofabric is shown in Fig.1. The fabric had the areal density ranged from 1.6 to 2.0 g/m^2, which is about 1% of prepreg areal density (about 150 g/m^2).

Fabrication of Panels and Test Specimens

Unidirectional (0°) AS4-3501-6 laminates were fabricated for tension, compression, short beam shear and Mode I fracture and delamination onset life tests. For each type of test one base (non-interleaved) panel and one interleaved panel by Nylon-66 nanofiber were made. Tension, compression and short beam shear panels had 10, 20 and 20 plies, respectively. The interleaved panels were made by placing one layer of nanofiber fabric over each of the prepreg plies except for the last ply (see Fig.2a). Fracture and fatigue delamination onset life panels were 20-ply thick and a Teflon film was placed between the 10th and 11th plies to create the initial delamination. The interleaved panel was made by placing two layers of nanofiber fabric between the 10th and 11th plies of prepreg (see Fig.2b). All the panels were made in autoclave as per the guidance provided by the prepreg manufacturer. The average areal density of a single layer of the Nylon-66 nanofabric was about 1.5 g/m^2. Calculated thickness (based on the assumed density of 1.14 g/cc for Nylon-66) of the interleave fabrics was 1.3 μm. Average thickness of tension, compression, short beam shear and fracture and fatigue onset life test panels was 1.40, 2.72, 2.77 and 2.70 mm, respectively, for base panels and was 1.41, 2.76, 2.77 and 2.70 mm, respectively, for interleaved panels. The maximum thickness increase due to interleaving was less than 1.5%.

Test

a. Tension : Longitudinal tension test was conducted according to ASTM Standard Test Method D3039/D 3039M-00. Test specimen dimensions were: length 254 mm (10 in), width 12.7 mm (0.5 in) and tab length 50.8 mm (2 in). Five base specimens and five interleaved specimens were tested. The test was conducted on an MTS test machine. Load, displacement and transverse strain measured by strain gage were recorded continuously till the specimen fractured. From this data, tensile modulus, strength, and Poisson's ratio were reduced as per the standard. Failure modes during initiation, progression, and final fracture were recorded. Specimen geometry, tensile strength, modulus, and Poisson's ratio of base and interleaved specimens are listed in Table-1. Average values and standard deviations (STD) are presented.

b. Compression : Longitudinal compression test was conducted according to ASTM Standard Test Method D3410. Test specimen dimensions were: length 140 mm (5.5 in), width 12.7 mm (0.5 in) and tab length 50.8 mm (2 in). Five base specimens and five interleaved specimens were tested. Load and displacement were recorded con-

Table-1 : Tension Test Results

Laminate	Specimen No.	Thickness, mm	Width, mm	Tensile Strength F_{1t}, MPa	Tensile Modulus E_{1t}, GPa	Poisson's Ratio ν_{12}
Base	T-NI-1	1.39	12.79	2,175	145	0.29
	T-NI-2	1.40	12.86	2,111	130	0.33
	T-NI-3	1.39	12,95	2,148	140	0.30
	T-NI-4	1.40	12.95	2,124	136	0.33
	T-NI-5	1.41	12.80	2,052	138	0.34
	Average	1.40	12.87	2,122	138	0.32
	STD	0.01	0.08	46	5.4	0.02
Interleaved	T-I-1	1.41	12.69	2,184	137	0.29
	T-I-2	1.40	12.95	2,178	137	0.33
	T-I-3	1.40	12.90	2,195	144	0.31
	T-I-4	1.42	12.90	2,084	137	0.31
	T-I-5	1.41	12.78	2,304	135	0.30
	Average	1.41	12.85	2,189	138	0.31
	STD	0.01	0.11	78	3.7	0.01

tinuously till the specimen fractured. From the failure load, compressive strength was calculated. Specimen geometry and compression strength of base and interleaved specimens are listed in Table-2. The average values and standard deviations are presented.

c. Short Beam Shear : Four point loaded short beam shear test [10] was conducted to measure the interlaminar shear strength (ILSS) of the material. The specimen width was 5.08 mm (0.2 in) and the span length was 11.43 mm (0.45 in). Five base and five interleaved specimens were tested. Failure load was recorded and the ILSS was calculated from the failure load using Eq.1.

$$F_{ILSS} = 0.75 \times \frac{P_m}{bh} \tag{1}$$

Where P_m is the maximum load, b is the specimen width and h is the specimen thickness. The edge of the specimen was polished before the test and was examined for failure modes using optical microscope after the test. Specimen dimensions, failure loads, and ILSS are listed in Table- 3.

Mode-I Fracture and Delamination Onset Life : The Mode I fracture test was conducted using a double cantilever beam (DCB) specimen of 230 mm long, 20 mm wide and 2.7 mm thick. The initial delamination length was about 50 mm. Fig.3 shows the specimen configuration and the loading. The fracture test was conducted according to ASTM Standard D5528. The test was carried out in a MTS test machine using a 880N (200 lb) load cell under displacement control at a constant cross-head rate of 1.3 mm/min. Load, cross-head displacement, and delamination length (a) were recorded continuously during the test.

The energy release rate G_I was calculated from the modified beam theory.

$$G_I = \frac{3P\delta}{2b\,(a + |\Delta|)} \tag{2}$$

where P is the load, δ is the load point displacement, b is the specimen width, a is the delamination length and Δ is the delamination length correction parameter for not perfectly built-in condition of the DCB. Value of Δ is established after the test. Maximum load and the associated deflection (δ) at the initial delamination length (a_0) was used to calculate the initiation fracture toughness G_{IC} of the material. For $a > a_0$, the G_I becomes G_R, fracture resistance. The G_R, once it becomes nearly constant with delamination propagation, is called G_{IR}.

The Mode I fatigue delamination growth onset life test was conducted according to ASTM D6115 to obtain the fatigue delamination onset life $N_{1\%}$, number of cycles for 1% compliance increase. The test specimens had the

Table-2 : Compression Test Results

Laminate	Specimen No.	Thickness, mm	Width, mm	Compressive Strength F1c, MPa
Base	C-NON-1	2.69	12.83	1,543
	C-NON-2	2.74	12.98	1,543
	C-NON-3	2.72	12.90	1,599
	C-NON-4	2.74	12.93	1,539
	C-NON-5	2.69	12.47	1,497
	Average	2.72	12.82	1,544
	STD	0.03	0.20	36
Interleaved	C-INT-1	2.74	12.88	1,544
	C-INT-2	2.74	12.47	1,492
	C-INT-3	2.77	12.60	1,481
	C-INT-4	2.77	12.62	1,578
	C-INT-5	2.77	12.70	1,559
	Average	2.76	12.65	1,531
	STD	0.01	0.15	42

Table-3 : Short Beam Shear Test Results

Laminate	Specimen No.	Thickness, mm	Width, mm	Span, mm	Failure Load, N	SBS Strength, MPa
Base	N-SBS-2	2.77	5.59	11.43	2,451	119
	N-SBS-3	2.82	5.84	11.43	2,629	120
	N-SBS-4	2.82	5.59	11.43	2,562	122
	N-SBS-5	2.74	5.59	11.43	2,611	128
	N-SBS-6	2.72	5.59	11.43	2,620	129
	Average	2.77	5.64	11.43	2,575	124
	STD	0.05	0.11	0	74	5
Interleaved	IT-SBS-1	2.82	5.59	11.43	2,664	127
	IT-SBS-2	2.77	5.59	11.43	2,745	133
	IT-SBS-3	2.74	5.54	11.43	2,475	122
	IT-SBS-4	2.77	5.56	11.43	2,708	132
	IT-SBS-5	2.77	5.59	11.43	2,717	132
	Average	2.77	5.57	11.43	2,662	129
	STD	0.03	0.02	0	34	3

same configuration as the fracture test. The test was conducted under displacement control with loading ratio $R = \delta_{Imin}/\delta_{Imax} = 0.3$. Tests were conducted for different values of G_{Imax}. Equivalent maximum displacement δ_{Imax} equation was derived using the beam theory and the specimen similarity approach as explained in Ref.17 and is given by:

$$\delta_{Imax} = \sqrt{\frac{G_{Imax}}{G_{IC}}}\left(\frac{a_0+\Delta}{a_{IC}+\Delta}\right)^2 \delta_{IC} \tag{3}$$

where a_0 is the initial delamination length for fatigue onset life test, a_{IC} is the initial delamination length of the fracture test used in this calculation, Δ is the delamination length correction parameter determined from the fracture test, and δ_{IC} is the critical load-point displacement when the delamination starts to grow in the fracture test. The test specimen was cycled until the compliance increased by 1%. The fatigue delamination onset life $N_{1\%}$ was determined for different values of G_{Imax} and from that data $G_{I\ Threshold}$ was determined. The $G_{I\ Threshold}$ is the G_{Imax} required to increase 1% compliance in one million load cycles.

Results and Discussion

a. Tension : Longitudinal tensile strength, modulus and major Poisson's ratio of base AS4/3501-6 composite were 2,122 MPa (STD=46 MPa), 138 GPa (STD=5.4GPa) and 3.2, respectively. Those for interleaved AS4/3501-6 composite were 2,189 MPa (STD=78 MPa), 138 GPa (STD=3.7 GPa) and 3.1, respectively. The properties difference between the base and interleaved lamintes is within the data scatter. Therefore, polymer nanofiber interleaving does not alter the inplane tensile modulus and strength. This is expected because the specimen thickness is not significantly changed by interleaving. Tensile strength and modulus results agree reasonably well with the literature data which has a large range [11] ($F_{1t} = 2{,}280 \pm 330$ MPa, $E_{1t} = 147 \pm 21.3$ GPa). Whereas Poisson's ratio is slightly higher than the literature data ($\nu_{12} = \ 2.7$).

b. Compression : Compression strengths for base and interleaved specimens were 1,544 ± 36 MPa and 1,531 ± 42 MPa, respectively. The strength difference between the laminates is within the data scatter. The two results agree with the literature [18] data (1,725 ± 250 MPa). Both base and interleaved types of specimens failed by fiber kinking. Fig.4 shows the typical failure mode. Because the polymer nanofabric interlayer thickness is about 1% of the ply thickness, both in-plane tensile and compressive properties remained unaltered.

c. Short Beam Shear : The interlaminar shear strength of interleaved AS4/3501-6 was 129 ± 3 MPa compared to 124 ± 5 MPa of base specimens. Polymer interleaving increased interlaminar shear strength by 4%. Fig.5 shows the delaminations between the layers. The delaminations occurred between the top and bottom loading points. Tilting of the fibers was because of the specimen deformation under the flexure loading.

d. Mode-I Fracture and Delamination Onset Life : The energy release rate (G_I) versus delamination extension (*da*) for base and interleaved AS4/3501-6 composites were plotted in Fig.6. Hollow symbols represent base specimens while solid symbols represent interleaved specimens. Different symbol types represent different test specimens (two for base and three for interleaved). The average initiation fracture toughness G_{IC} of base and interleaved AS4/3501-6 was 84 J/m^2 and 212 J/m^2, respectively, about 150% increase. For base AS4/3501-6, the fracture resistance increased with the delamination propagation and reached a constant value of about 154 J/m^2 after 25 mm of delamination propagation. On the other hand, interleaved AS4/3501-6's resistance decreased with delamination growth and leveled off at about 201 J/m^2, an increase of about 30% over the base laminate. These percentage increases of G_{IC} and G_{IR} are of the same order as those of T800H/3900-2 when compared with its non-interleaved T800H/3631 composite but with no penalty on thickness increase [12-15]. Fig.7 shows the SEM image of the fracture morphology of the nanofabric interleaved AS4/3501-6 specimen. Stretching, separation, and breakage of Nylon-66 nanofibers are shown on the delaminated surface. These are the case of reasons for toughness enhancement.

Two base specimens (G_{Imax} = 67 and 42 J/m^2) and four nanofabric interleaved specimens (G_{Imax} = 170, 106 and 53 J/m^2) were tested for delamination onset lives for R=0.3. The stress ratio, here the G ratio of 0.3 was chosen to avoid contact of delaminated surfaces. Fig.8 shows G_{Imax} versus delamination onset life $N_{1/\%}$ for the two material systems. Notice a wide separation between the two curves that signifies the impact of nanofabric interleaving on the fatigue onset life. Onset threshold $G_{I\ Threshold}$ (G_{Imax} at N = 10^6 cycles) values of base and nanofabric interleaved AS4/3501-6 are 30 J/m^2 and 50 J/m^2, respectively, which reflect an increase of 67% threshold G.

Concluding Remarks

This research was to demonstrate the concept of polymer nanofiber interleaving to enhance the toughness of fiber reinforced carbon/epoxy composite laminates. The Nylon-66 nanofiber was produced by electrospinning and Nylon-66 was chosen for its compatibility with epoxy. In-plane tension, compression, interlaminar shear, and Mode I fracture and fatigue onset life properties were measured. Results demonstrated that by adding 1% weight of nanofibers in the form of interlayer the in-plane tension and compression properties (strength, modulus, and Poisson's ratio) of interleaved composites practically unaltered. Whereas it increased interlaminar shear strength by 4%, fracture toughness by 150% and fatigue threshold energy release rate by 67%.

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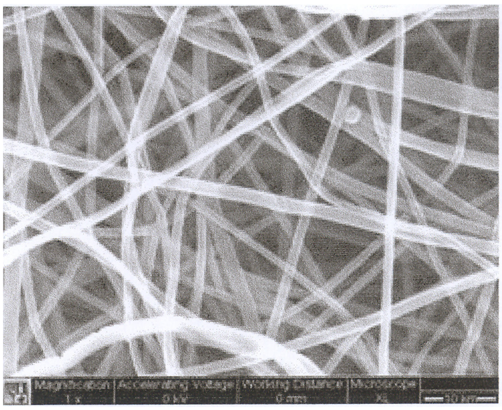


Fig.1 SEM Image of Electrospun Nylon-66 Fabric

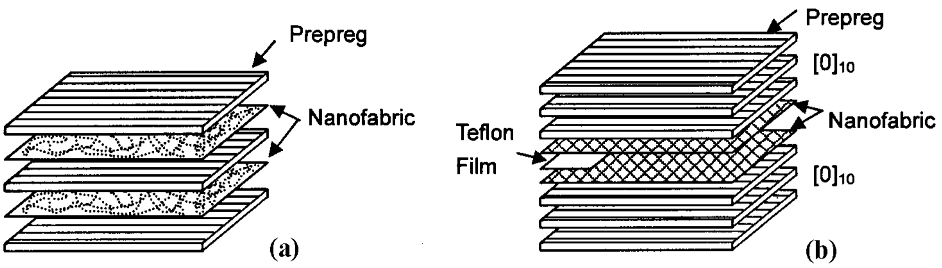


Fig.2 Schematic of Interleaving Procedure (a) Tension, Compression and Short Beam Shear Panel (b) Fracture Test Panel

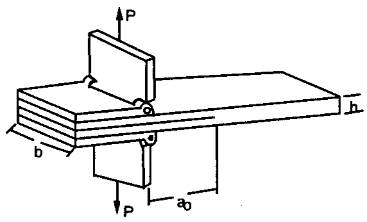


Fig.3 Fracture and Fatigue Onset Tests Specimen

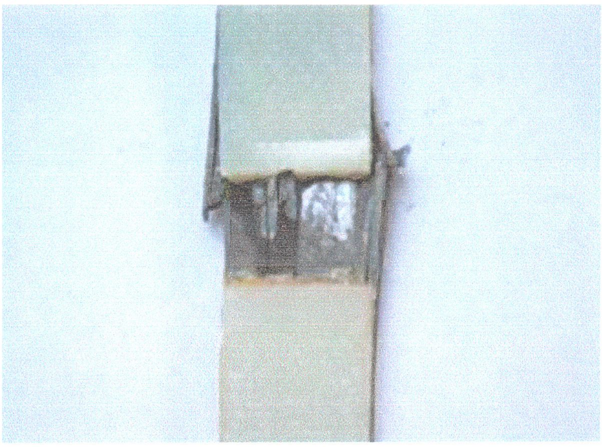
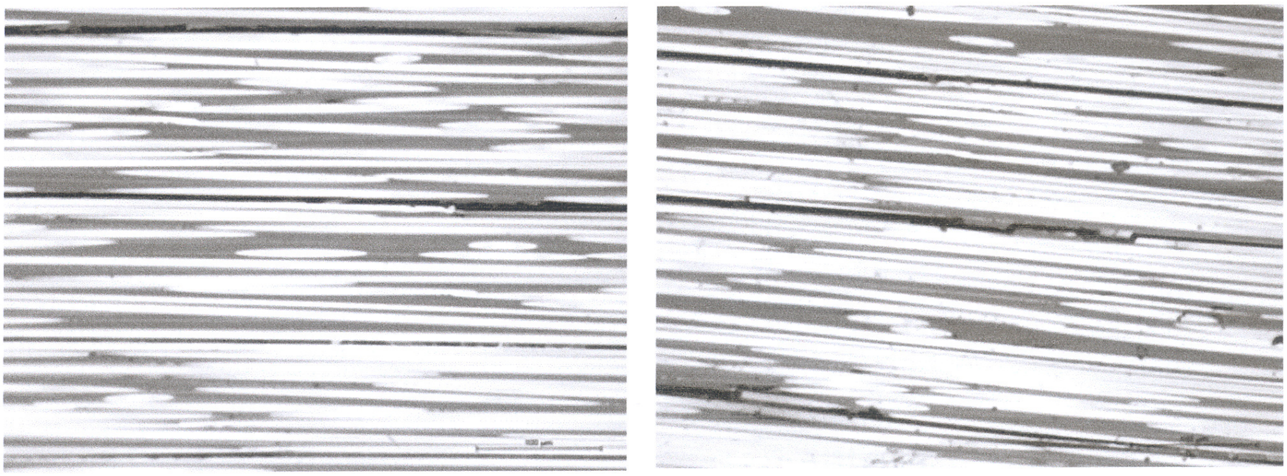


Fig.4 Failure Mode of Compression



(a) (b)

Fig.5 Interlaminar Shear Failure Mode (a) Base Specimen (b) Interleaved Specimen

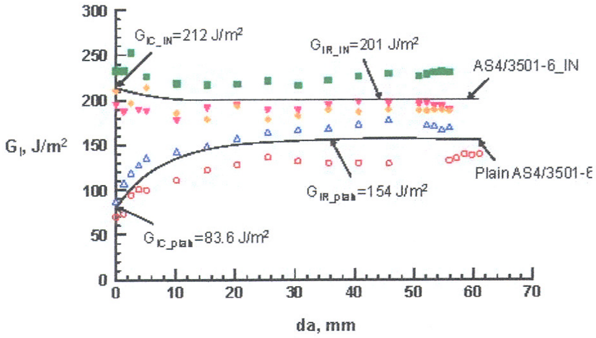


Fig.6 Fracture Toughness and Resistance of Base and Interleaved AS4/3501-6 Composites

Fig.7 SEM Image of the Fracture Surface of the Nanofabric Interleaved AS4/3501-6 Specimen

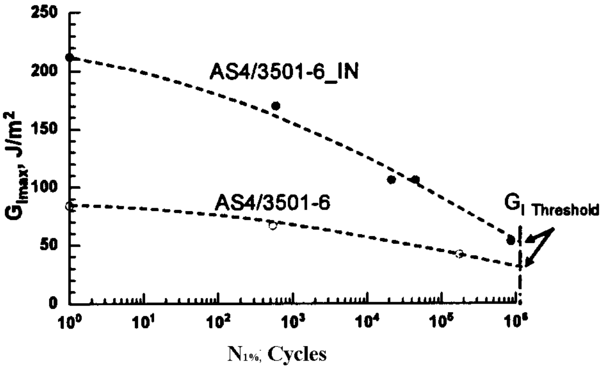


Fig.8 G_{Imax} Versus Fatigue Onset Life for Base and Interleaved AS4/3501-6 Composites