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Interlaminar shear strength of multi-walled carbon nanotube and carbon fiber reinforced, epoxy - matrix hybrid composite

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Abstract

Studies have been carried out to evaluate interlaminar shear behaviour of carbon nanotube and carbon fiber reinforced, epoxy matrix hybrid composite. Short beam shear (SBS) tests were conducted to characterize the influence of fiber orientation in the interlaminar shear strength (ILSS). Experimental details, specimen configuration, data acquisition and processing are presented in detail. The present study reveals the importance of alignment of fiber and its effects on ILSS properties and nature of deformation under shear loading conditions. The results show that the interlaminar shear strength of the hybrid composite is significantly higher in longitudinal orientation as compared to the same in transverse orientation; and more importantly show that such anisotropy is of an order of magnitude and higher.

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Keywords: Hybrid Composite; short beam shear test; interlaminar shear strength; multi-walled carbon nanotubes

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1. Introduction

Continuous fiber – reinforced polymer matrix composites have been used successfully in many industries such as aerospace, automobile, marine, military, etc., their interlaminar shear strength (ILSS) is usually one of the most common limiting design characteristic. Improving ILSS has long been an important goal in the fiber reinforced composites field, and to this end different approaches have been tried. Some of the more effective attempts have been weaving fibers in the thickness direction or using Z-pins to connect the laminate have been carried out by Byrd et al. (2006), Jain et al. (1994), Sharma et al. (1997) and Zhang et al. (2006). Abali et al. (2003) have reported that these techniques are labor intensive and require additional manufacturing processes that can greatly increase the cost of the resulting components.

Hsiao et al. (2003), Zhou et al. (2006), Yokozeki et al. (2006) and Zhu et al. (2006) have explored other ways to improve the ILSS by resorting to the reinforcement of nanoparticles and carbon nanotubes. Siddiqui et al. (2006) found that adding organoclay nanoparticles increases both crack growth resistance and fracture toughness in carbon fiber reinforced polymer matrix composites and hence it is most desirable. Zhu et al. (2005) proposed an innovative way to create CNT/fiber/polymer composites by coating single walled carbon nanotubes (SWNTs) on the surface of glass fibers before filling the mold with vinyl ester and thereby obtained a 35% enhancement in ILSS. Yokozeki et al. (2006) found that carbon fiber reinforced composites can benefit from dispersion of cup-stacked carbon nanotubes (CSCNTs) between fiber mats and these can delay the onset of matrix cracking. Fan et al. (2008) found that the addition of oxidized multi-walled carbon nanotubes (OMWNTs) improved the ILSS in glass fiber reinforced epoxy composites made by both the distribution-media-assisted vacuum assisted resin transfer molding (VARTM) and injection and double vacuum assisted resin transfer molding (IDVARTM) methods. However, the studies of the effect of directionality of fibers on the ILSS properties of hybrid composites have not been performed so far.

The results from the present developmental work have shown an encouraging path toward CNT application in fiber reinforced composites. This paper aimed to study the ILSS properties of the unidirectional amino multi-walled carbon nanotube (AMWCNT) and carbon fiber reinforced, epoxy matrix hybrid composite in detail in order to determine the influence of directionality of fibers. The obtained results are rationalized based on materials' characteristics and mode of failure.

2. Experimental

2.1. Materials

An epoxy resin based on diglycidyl ether of bisphenol-A (DGEBA) and diethyl toluene diamene (DETDA) hardener were used as matrix system. T-700 carbon fiber tows with ultimate tensile strength of 4800 MPa and amino functionalized multi walled carbon nanotubes were used as reinforcing materials. The tube diameter ranges from 20 to 30 nm and the tube lengths ranges from 2- 4 μm. The amount of CNTs dispersed in the epoxy resin was kept constant to 0.5 wt. % (weight %). Recent research shows that the tensile strength values were improved significantly by the addition of MWNTs to epoxy. However, the improvement in the tensile strength occurred until MWNTs was added up to 0.5 wt%. This is because in higher contents of filler, there are severe and localized agglomerations within the nanocomposite. The presence of voids produced during stirring the epoxy/MWNTs suspension with the hardener, also decreases the mechanical properties. Ayatollahi et al. (2011) have performed experiments on nanocomposites and have reported that by adding more MWNTs the viscosity of the mixture increases which makes the degassing of mixture more difficult. The synthesis route adopted in the present study was via a conventional hand layup technique as outlined elaborately by Chandra Shekar et al. (2013). The densification and curing process is carried in a muffle furnace in three heat treatment cycles. The final material obtained had a thickness of 3.4 mm, and density of 1.56 g/cc, with volume fiber fraction of 82 %.The microstructure of hybrid composite was observed by using scanning electron microscope. Table 1 shows the physical properties of the hybrid composite.

Table 1. Physical properties of the hybrid composite.

S. No	Physical Property	Value obtained
1	Density	1.56 g/cc
2	Fibre volume fraction	82.05%
3	Young's modulus (Longitudinal)	16.31 GPa
4	Young's modulus (Transverse)	15.9 GPa

2.2. *Specimen Preparation*

Short beam shear test 3- point bend specimens were machined in both longitudinal and transverse orientations and pertain to the geometry of the specimen as described in ASTM D 2344. Short beam shear test 3- point bend loading were conducted on specimens of width (w) of 10 mm, thickness (t) of 3.40 mm and length of 50 mm (with an included span length equivalent to 6 times the thickness).

2.3. *Interlaminar shear strength characterization*

When the transverse shear load experienced by a laminated hybrid composite exceeds the interlaminar shear strength (ILSS), a delamination failure will occur between the layers of reinforcing fibers. To measure the ILSS of a composite directly, a pattern of pure shear stress should be generated between laminae to induce an interlaminar shear failure. A number of different tests such as short beam shear test (SBS), curved-double-cantilever-beam (CDCB), compression shear test (CST) have been developed for the purpose of characterizing the ILSS. Comparing the available methods, the short beam shear method is the most simplest and therefore most commonly used in practice. Based on the classical beam theory, it causes transverse shear failure through three point bending as shown in Figure 1.

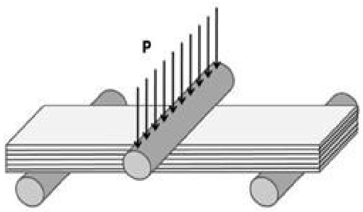

Fig.1. Short beam shear (Three point bending) testing.

The short beam shear test is designed to generate interlaminar shear indirectly through bending and is the most popular method to characterize the interlaminar shear strength of unidirectional, fiber – reinforced composites (ASTM D 2344). As shown in Figure 1, the specimen is placed on two cylindrical supports and a cylindrical head is moved down to apply a force at the center and generate an increasing transverse load until the first failure is recorded. The load at failure is then used to determine the interlaminar shear strength of the composite. The interlaminar shear strength was calculated in two orientations namely longitudinal orientation (fibers perpendicular to the direction of crack propagation) and transverse orientation (fibers are aligned parallel to the propagation of the crack).

3. Results and Discussion

3.1. Microstructure

The arrangement of carbon fibers was observed using scanning electron microscope (SEM) before testing of hybrid composite. The carbon fibers were found to be continuous throughout the matrix aligned unidirectionally. The average diameter of the carbon fiber was 6 µm (Figure 2a). Fibers were physically distinguishable and were not found bonded into one another in spite of three heat treatment cycles. Figure 2b clearly shows that there are absolutely no traces of damage in the form of any breakage among fibers.

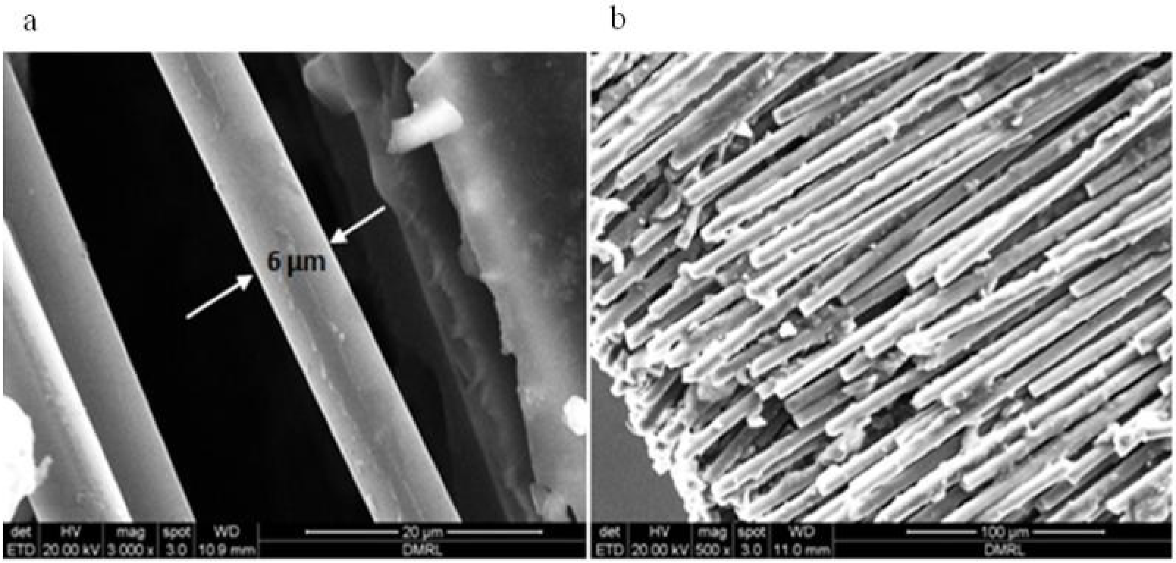

Fig. 2. SEM micrographs of carbon fibers in the hybrid composite (a) Single carbon fibre; (b) Carbon fiber bundles.

3.2. Interlaminar shear strength (ILSS)

The determination of ILSS using short beam shear testing is based on classical (Bernoulli-Euler) beam theory. The failure mechanisms during short beam 3-point bending are a combination of compression, tensile and shear deformations. Initiation sequence of any of these failure mechanisms depends on the state of a sample, for example, properties of the matrix (brittle or tough), porosity in the laminate, fiber volume fraction, etc. In the present case addition of CNTs in to matrix brings in an additional parameter. All the ILSS tests were conducted as per the ASTM D 2344 using universal testing machine (United, Model STM 50 KN, USA) with the help of a self articulating 3 point bend fixture. The tests were conducted at ambient temperature and in laboratory air atmosphere. ILSS tests were carried out under ramp control speed of 2 mm/min. The load versus displacement values were recorded as per ASTM standard D 2344. For a beam of rectangular cross section loaded in three-point bending, the maximum interlaminar shear stress occurs at the mid thickness of the beam between the center and end supports and it is given as:

$$\mathrm{ILSS} = 0.75\left(\frac{P_b}{wt}\right) \quad (1),$$

where, P_b= breaking load, w and t are width and thickness of the specimens respectively. A minimum of five specimens were tested in each orientation and for the sake of clarity, only one typical specimen load-displacement data set is shown in each of the cases i.e., longitudinal and transverse orientations.

3.2.1. ILSS properties and effect of fiber orientation on ILSS of hybrid composite

3.2.1.1. Longitudinal orientation

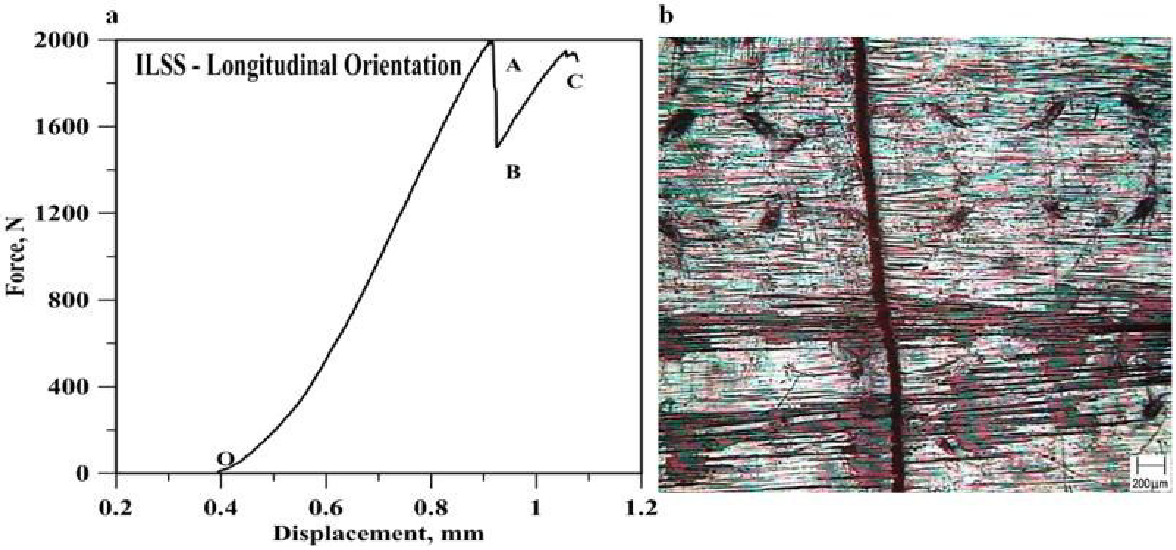

Fig. 3. (a) Load vs. displacement curve for a Short beam shear (3-point bend loading) specimen in longitudinal orientation; (b) Crack path observation after ILSS testing in longitudinal orientation.

The typical variation of load versus displacement for one of the specimens in longitudinal orientation is shown in Figure 3a. As stated earlier, the ILSS values for the hybrid composite, evaluated for longitudinal direction are obtained from testing of a minimum of five specimens and the data are included in Table 2. It is clearly seen from the Table 2 that the material possesses excellent interlaminar shear strength in the longitudinal orientation with an average value of 56 ± 9 MPa. The excellent ILSS values in case of the present hybrid composites can be attributed to the presence of light weight carbon fibers and also the particulate reinforcement- the amino functionalized multi walled carbon nanotubes. As can be seen from the data in Figure 3a, the load rises gradually (OA) and then drops showing a distinct occurrence of major fiber bundle failure (at A) and subsequent crack arrest (at B). The gradual increase in the elastic zone (in OA region) is due to the presence of fibers perpendicular to the direction of crack propagation and also due to the AMWCNTs, which take the load completely. This is followed by a sharp and steep fall in the stress value (at A) with a constant displacement. This followed by a small increase in the load (BC) after this, steep decrease in load occurs until final fracture resulting in complete fracture/separation. Table 2 gives the overall information regarding the testing carried out in longitudinal orientation. Figure 3b shows the photo micrograph of the fractured surface when the direction of crack propagation is across the length of the specimen. Resin micro cracks were also observed along the length of crack. The calculated ILSS therefore represented ultimate failure under a combination of resin micro cracks and interlaminar shear.

Table 2. Dimensions of specimens used and the values of ILSS of the hybrid composite in longitudinal orientation.

Sl. No	Specimen	Width, mm	Thickness, mm	Max applied Load, N	ILSS, MPa
1	ILSS-L1	9.90	3.20	1996	47.26
2	ILSS-L2	10.04	3.30	2533	57.33
3	ILSS-L3	9.99	3.26	2735	62.98
4	ILSS-L4	10.10	3.20	2257	52.38
5	ILSS-L5	10.00	3.28	2051	46.89
6	ILSS-L6	10.04	3.19	2937	68.77

3.2.1.2. Transverse orientation

Using the procedure similar to the one adopted for longitudinal direction, the ILSS values for the hybrid composite in transverse direction too are determined. The data in Table 3 show that hybrid composite in transverse orientation has ILSS that is significantly lower (8 – 9 %) when compared to the longitudinal direction. The average ILSS obtained in this orientation is approximately 5 ± 0.75 MPa. This is because fibers are loaded perpendicular to their long axis, when tested in transverse direction; and the fibers are inherently susceptible to biaxial loading across the fiber loading direction. In this case, matrix (epoxy and AMWCNTs) takes the entire load and attained a critical value. Perhaps, the ILSS values would even to a lower, if CNT reinforcement is not incorporated which hinders the crack propagation.

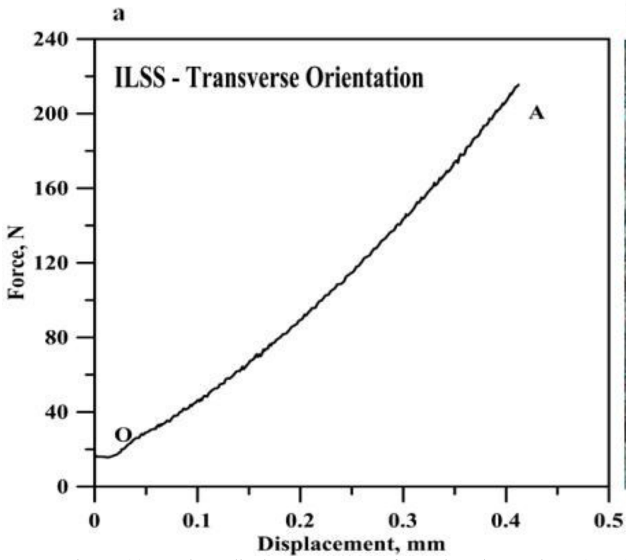

Fig. 4. (a) Load vs. displacement curve for a short beam shear (3-point bend loading) specimen in transverse orientation; (b) Crack path observation after ILSS testing in transverse orientation.

The load-displacement variation obtained, for the transverse alignment of fibers under 3-point bend loading shown in Figure 4a, exhibits brittle fracture that is indicated by the sudden drop in the load at "A" immediately after the attainment of peak stress. It is observed that the maximum average load of this composite in transverse orientation is 210 ± 32 N approximately which corresponds to an ILSS of 5 ± 0.75 MPa. Table 3 gives the detailed account of testing carried out in this orientation. Figure 4b shows photomicrograph image of specimen tested in SBS and shows the propagation of crack across the length of specimen. It can be easily seen from the photo micrograph that the fracture is very sharp without much lateral displacements or fracture of fibers.

Table 3. Dimensions of specimens used and the values of ILSS of the hybrid composite in transverse orientation.

Sl. No	Specimen	Width, mm	Thickness, mm	Max applied Load, N	ILSS, MPa
1	ILSS-T1	9.52	3.24	215	5.24
2	ILSS-T2	10.40	3.25	255	5.65
3	ILSS-T3	10.25	3.25	185	4.16
4	ILSS-T4	10.19	3.27	174	3.92
5	ILSS-T5	9.73	3.31	222	5.16

3.3. Directionality in strength properties

From the above tests conducted and results obtained, it is clear that a hybrid composite such as amino multi-walled carbon nanotube and carbon fiber reinforced, epoxy matrix material has significantly higher interlaminar shear strength in longitudinal orientation of fibers compared to transverse orientation of fibers. This is clearly reflected in the load vs. displacement data of the material when plotted together on the same scale for both the orientations (Figure 5).

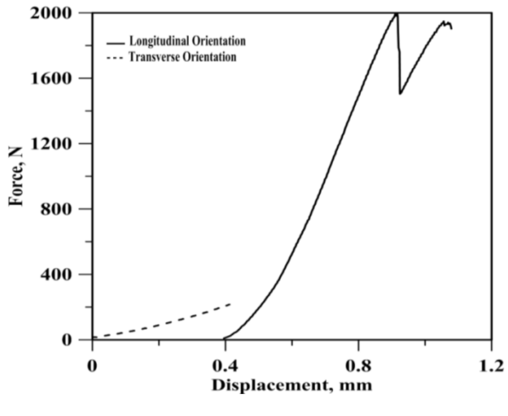

Fig. 5. Load vs. displacement data for ILSS (3-point bend loading) test for longitudinal and transverse orientations.

Though a similarity can be observed in the nature of load-displacement variation for the material in two different test directions, there is a huge disparity in the stress bearing capacities. The hybrid composite showed an isotropic behaviour with almost same value of Young's modulus in both the orientations (17 GPa; please see data in Table 1). On the other hand, as pointed earlier the ILSS is vastly anisotropic. This is because the fact that fibers have a dominant role to play. Fibers predominantly resist the crack propagation in longitudinal orientation, while the matrix provides the maximum resistance to final fracture of the material when load is applied in the transverse orientation.

Conclusion and Technological implications

The interlaminar shear strength of the hybrid composite is significantly higher (10 times or nearly 1000 %) in longitudinal orientation as compared to the same in transverse orientation. Based on the results obtained for the present study that the hybrid composite material should be designed, so that the major stress axis is in the longitudinal orientation and should avoid loading in the transverse orientation as much as possible. If the design is based on lowest ILSS values the true potential of hybrid composite cannot be achieved. This is because the transverse ILSS is an order of magnitude lower. With these properties obtained from the hybrid composite, it is clearly suitable for structural applications in the aeronautical and aerospace industry, where the exceptionally high values of longitudinal interlaminar shear strength can be exploited. Hence the present hybrid composite possesses high anisotropic ILSS properties.

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