A TOUGHENING AND CONDUCTIVE APPROACH FOR HYBRID COMPOSITES WITH NON-WOVEN CARBON TISSUE INTERLEAVES

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ABSTRACT

To enhance both delamination resistance and electrical conductivity of CFRP laminates，the non-woven carbon tissues (NWCT) composed of short chopped carbon fibers were prepared as interleaves with a simple surfactant-assistant dispersion and filtration method. The end notch flexural (ENF) and flexural mechanical test indicate the incorporation of NWCT interleave can improve mode II fracture toughness by 97% without compromising of flexural modulus and flexural strength. More interestingly, comparing with the baseline composites, the electrical conductivity of the interleaved composites exhibited an enhancement of 96% in the in-plane and 82% in the through-the-thickness direction, respectively. Microscopy analysis of carbon tissue interleaving area in the laminate indicates that the chopped short carbon fibers can form into a 3D network with more fibers aligned along through-the-thickness direction, resulting in significantly improvement of electric conductivity in through-the-thickness direction. The SEM observation reveals that fiber pullout and epoxy plastic deformation are the major toughening mechanism involved in the delamination leading to the toughness improvement.

1 INTRODUCTION

Composite laminates have been widely used in weight-critical structures, such as aircraft, spacecraft, racing car, etc, due to excellent mass-specific mechanical properties. Unfortunately, poor delamination toughness [1] in piles of composite laminate has become the important limiting factor in practical structure application. The extensive methods were obtained for improving the delamination toughness of CFRPs composites, including the techniques of the stitching [2-3], Z-pin[4-6], the surface modification of carbon fabrics with CNTs[7-9] and interleaving [10-19]. A simple and low-cost method to toughen laminate composites is to utilize the chopped Kevlar fiber in the mid-plane of laminates developed by Hu[20-22], where the manually spread chopped Kevlar fibers with around 2-15mm in length in the mid-plane can obviously enhance both the interlaminar fracture toughness of the composites, the toughened mechanisms mainly come from the bridging effect of the random distributed chopped fibers which increase the energy dissipation as the crack propagation [22], however, the data for the laminates with short Kevlar fibers shows a wider scatter due to the uneven distribution of the Kevlar fibers probably come from the manually spread method. Recently various polymer films made of electrospun fibers have been utilized as interleaving materials in CFRPs [10-12], such as poly (ε-caprolactone)(PCL), polyacrylonitrile(PAN), polyvinylidene fluoride(PVDF), polyetherketone(PEK), and polysulfone, and resulted in varied success in the toughness improvement of the composites. For instance, electrospun PCL [10] and PEK[11] films were found to produce composite with enhanced mode I interlaminar fracture toughness(GIC), stable crack growth and maintained flexural strength and modulus. However, the incorporation of the polymer fibers will lead to the reduction of the electrical conductivity of the laminates and possible decrease in-plane strength and modulus of the laminates due to the high electrical insulation and weak mechanical properties of the polymer material in nature. Therefore, carbon tissue made of carbon fibers or CNT films was recently studied as the interleaving materials. With the help of the vaccum assisted infiltration, partially cured functionalized CNF film (Bulky paper) was utilized the interleaving materials for CFRPs and 104% increase of Mode II fracture toughness was achieved [14]. Comparing with the high price and complex fabrication procedure including the chemical functionalization of the bucky paper, the carbon tissue directly made of carbon fibers should be more desirable for the practical industry application. The non-woven carbon tissue (NWCT) was introduced into the interleaved reinforcement for the laminates composites by Lee et al [15-18]. The nonwoven carbon fiber tissue (15mm long) can significantly increase mode II interlaminar fracture toughness of the laminated CFRP composites by over 200% [15], without compromise the in-plane properties including stiffness, strength and fatigue life [17]. However, no significant change was found for the Mode I interlaminar fracture toughness using this interleave material the laminates [16], even with the modification of the CNT on the tissue[19]. Our recent study [23] have shown that NWCT interleave composed of shorter carbon fibers (0.8mm in length) with optimum density (7.8mg/cm^2) of the NWCT interleave, achieving a significant mode I fracture toughness enhancement of 99% compared with that of the baseline laminate, indicating the superior toughening ability of short carbon fibers in laminate on mode I fracture toughness.

Herein, the purpose of this paper is to utilize NWCT interleave composed of shorter carbon fibers (0.8 mm in length) to extend our investigation on interlaminar mode II fracture toughness, flexural mechanical properties and electrical conductivity improvement of laminated composites. The toughed mechanism for non-woven carbon fiber tissue is discussed based on the microscopy observation.

2 EXPERIMENTAL WORK

2.1 Materials

Materials used in this study were woven carbon fibres (168058ITL supplied by Inter-Turbine Advanced logistics Pty Ltd, Australia) for the CFRPs laminates composites and NWCF interleave preparation, the epoxy resin system including Araldite-F (diglycidyl ether of bisphenol A, DGEBA) and piperidine, supplied by Sigma-Aldrich, and surfactant (cellulose, supplied by Sigma-Aldrich).

2.2 NWCT fabrication

The woven carbon fibres were manually chopped into short carbon fibers (SCFs) with around 0.8mm in length. As shown in Fig.1, the aqueous dispersion containing chopped carbon fiber, and surfactant was stirred for 60min to prepare the uniform dispersion of chopped carbon fiber in the solution. After filtration of the dispersion and rinsed with distilled water to remove the residual surfactant, the final NWCT was obtained. The density of prepared NWCT interleave is 7.8mg/cm^2 corresponding to the thickness of 150μm in thickness.

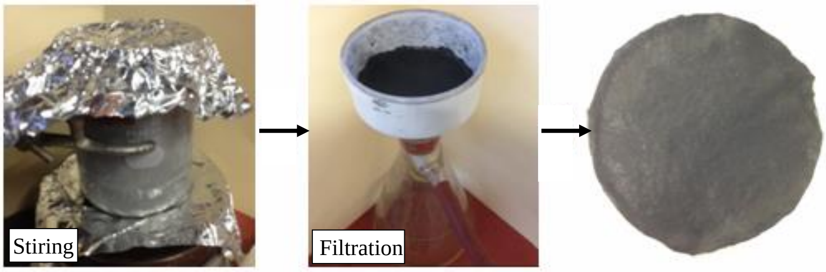


Figure 1 The procedure of preparing non-woven carbon tissue

2.3 Laminates preparation

The CFRPs laminates composites were fabricated from 16 plies of plain woven carbon fibres and neat epoxy by the hand lay-up method with the method used before [24]. A 0.2mm thick Kapton polyimide film and the prepared NWCT were inserted at the mid-plane of the laminates to serve as the pre-crack and interleave respectively to fabricate the sample for the interlaminar fracture toughness tests, illustrated as Figure 2.The laminates were wrapped with bleeders and release film within a vacuum bag, and first vacuumed in a chamber for 20min followed by curing in a hot-press at 120ºC for 16h. A pressure of 250kPa was applied during curing to maintain a uniform laminate thickness and a constant fibre volume fraction, which were 3.1mm and 60±2%, respectively. ENF (End-Notched Flexure) specimens were finally cut from the square panels by a wet-jet diamond saw. In addition, two-ply laminates interleaved with NWCT, were also prepared with the same method above for the electric conductivity measurement.

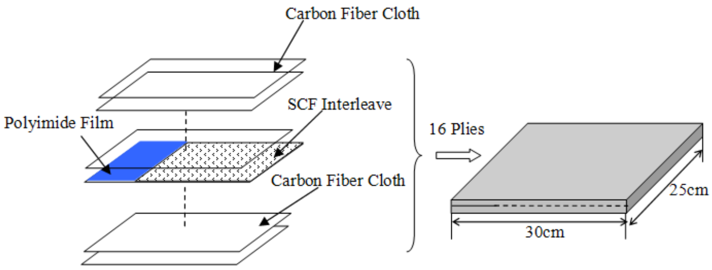


Figure 2 The illustration of the configuration of NWCT interleaved laminates for the interlaminar fracture toughness tests

2.4 Experimental procedure

All the mechanical properties tests were performed on an Instron 5567 machine. Standard 3-point ENF tests were performed in an Instron 5567 machine according to the Protocaol for Interlaminar fracture Testing No.2(1992)[25]. Fig 3(a) shows the geometry and dimensions of the ENF sample. The initial crack length *a* is 25mm and a/L=0.5. The crack mouth opening displacement rate was 2mm/min. At least 4 samples were tested for each matrix system and their load-displacement curves were recorded. The interlaminar toughness G_{IIC} , was calculated according to the protocol [25] by:

$$G_{IIC} = \frac{9a^2\delta P}{2b(2L^3+3a^3)} \cdot \frac{1-0.6099\times(\delta/L)^2}{1+0.3766\times(\delta/L)^2} \tag{1}$$

Where δ and P are displacement and maximum force recorded at the load-point at fracture.

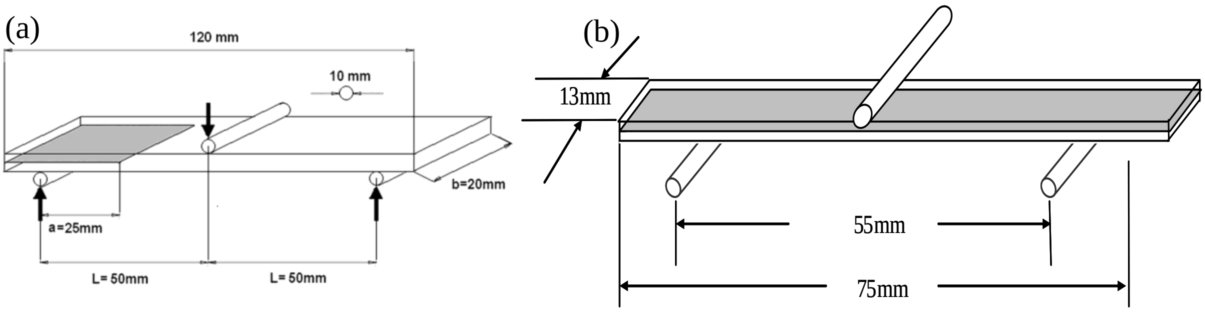


Figure 3 The specimen geometry for (a) ENF ; (b) Flexture specimen

The flexural properties of the CFRP laminate composites were determined from the three point bending test according to ASTM D790[26]. Rectangular specimens of 75mm long *13mm wide*3.3mm thick were loaded with a span of 55mm at a crosshead speed of 1.4mm/min, as shown in Figure 3(b). Five specimens were tested for each set of conditions. The conductivity of the interleaved laminates in both in-plane and through-the-thickness direction were measure with a CHI electrochemical workstation. To improve the electrical contact, silver paste was applied on certain sides of the samples.

2.5 Microstructure analysis

The dispersion of NWCT interleave was observed by the optical microscopy (OM).The fracture surface of the samples was coated with a thin gold layer and their morphologies were studied by SEM (Zeiss ULTRA Plus SEM) at an accelerated voltage of 2kV.

3 RESULTS AND DISSCUSSION

3.1 The morphology of chopped carbon fibers

The chopped SCFs can be well dispersed in the solution in the presence of the surfactant. After filtration, NWCTs composed of fibers was obtained, as shown in Fig 4(a). It can be found that the chopped carbon fibers are uniformly and randomly distributed in the NWCT and no fiber bundles/aggregation was observed, indicating that the original fiber bundles in the plain woven fabrics were exfoliated by the present nonionic surfactant in the solution and single fibers were well separated from each other. It is believed that the surfactant play an important role as stabilizer through the non covalent polymer wrapping to prevent the formation of bundles or aggregation, just like their role in the preparation of the stable carbon nanotube dispersions. The mean length SCFs is 0.8(±0.2mm) according to the statistics of 100 carbon fibers.

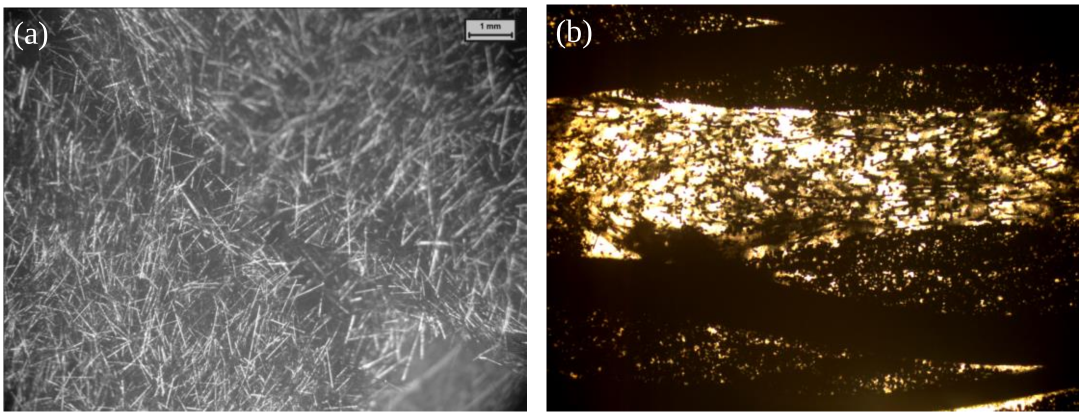


Fig.4 The optical images of (a) NWCT made of chopped carbon fibers; (b) the mid-layer of laminates interleaved with NWCTs

The NWCTs made of short carbon fibers(SCFs) were directly used as interleaves in the CFRP laminates. Fig 4(b) shows the dispersion of chopped carbon fibers in the mid-layer of laminate. For the NWCT interleaves made of SCFs, it can be seen that most SCFs are randomly dispersed and formed as the three-dimensional interwoven network structure, which would help to prevent the crack propagation efficiently and construct the conductive network in mid-ply of laminate. In addition, the SCFs enlarge the distance of adjacent layer of laminate, which help to increase the plastic zone of crack tip.

3.2 Interlaminar fracture toughness of NWCT-composites.

Since the optimum density of NWCT interleaves for the mode I fracture toughness improvement is 7.8mg/cm^2 [23], the NWCTs with the same density was prepared for the study on their interleaving effect on mode II fracture toughness of the laminate. Fig 5 compares the mode II fracture toughness G_{IIC} of pure CFRPs and those interleaved with NWCTs made of fibers. Notably, the incorporation of NWCTs can siginificantly increase the mode II fracture toughness of CFRPs laminates, which is increased by 97% for NWCT interleaves made of SCFs respectively. In fact, the G_{IIC} was even increased by more than 200% by using NWCTs made of longer fibers with length of 15mm [15], which indicated that the NWCTs with longer fiber length have better effect on the Mode II fracture toughness enhancement.

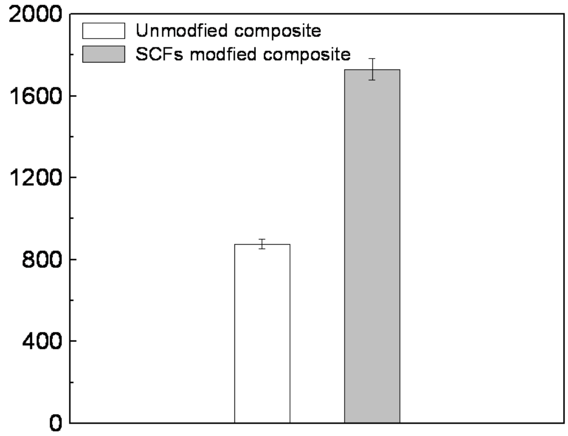


Fig 5. The G_{IIC} of NWCT interleaved CFRPs laminates

3.3 Flexural properties of short chopped carbon fiber reinforced laminate

Fig 6 shows the flexural properties of laminate interleaved with NWCT with density of 7.8mg/cm2, where the flexural strength and modulus of laminate were increased by 2.5% and 12% over the baseline composite respectively. The improvement of the flexural strength of laminate is owing to the stress transfer between the matrix and SCFs in the mid-layer.

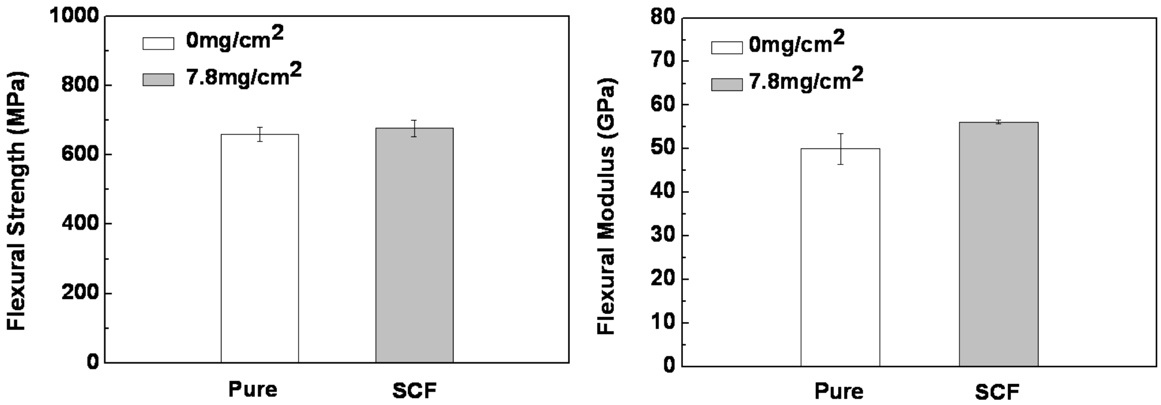
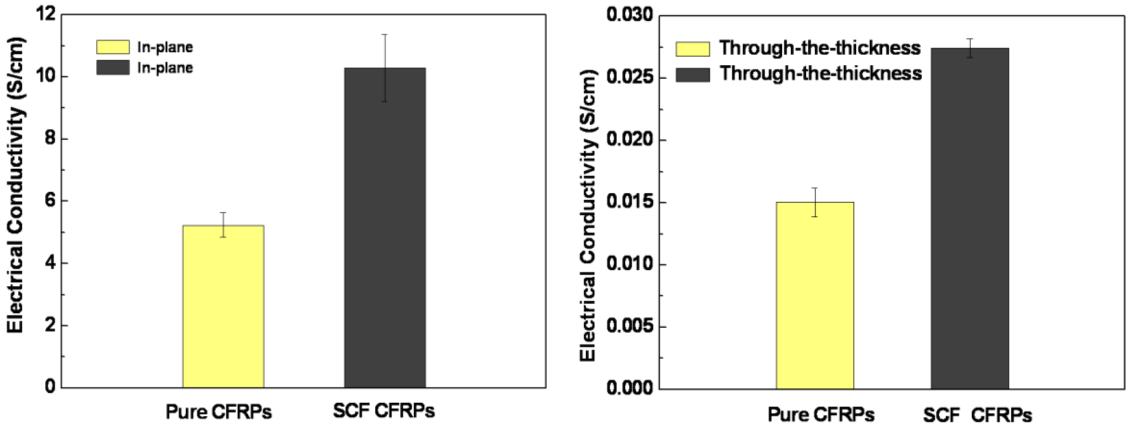


Fig. 6 Flexural properties of short chopped carbon fiber reinforced laminate

3.4 The conductivity of the NWCT modified laminates

Fig 7 shows the electrical conductivities of the laminates interleaved with NWCTs made of short fibers. The carbon fabric plies used in the composites is 0º/90º plain woven fibrics, which make the electrical conductivity of the composite quite different in the direction of in-plane and through-the-thickness of the laminates. The in-plane electrical conductivities were two magnitude orders higher than those in through-the-thickness direction, as shown in Fig 7 (a) and (b). Such huge difference mainly results from the laminated structure of the CFRPs composite. While continuous carbon fibers of the carbon fabrics are aligned along in-plane direction and directly build up the conductive network in this direction, the highly resistive epoxy resin-rich area is always located between carbon fabric layers, resulting in the reduced electrical conductivity in the through-the-thickness direction. Compared with the baseline CFRPs laminates, after the incorporation of NWCTs, the in-plane electrical conductivity was increased by over 96% (Fig 7(a)), while the through-the-thickness electrical conductivity was increased by over 82% (Fig 7(b)). The increase in electrical conductivity is owing to the formation of electrical conductive networks of the chopped carbon fibers in the matrix between fabrics plies. It can be seen that the improvement on conductivity in through-the-thickness direction was comparable with that in the in-plane direction. This can be attributed to the formation of the interlaminar 3D conductive network made of the SCFs in NWCTs.



(a) The conductivity in-plane (b) The conductivity in-thickness

Fig.7 The electrical conductivity of the laminates in different direction.

3.5 Toughening mechanism discussion

An overview of the Mode II delamination surface of a specimen interleaved with NWCTs is shown in Fig.8. The chopped carbon fibers are those without any fixed pattern, where many of them have been torn away from their original positions, and they were pulled out due to shear traction stress during delamination with evidence of long sheath left on the fracture surface. It can also be observed that the chopped carbon fibers show the crack bridging effect with evidence of fractured carbon fibers. The pullout or fracture behavior of chopped carbon fibers can cause the matrix deformation finally leading to rougher fracture surface and improved dissipated fracture energy.

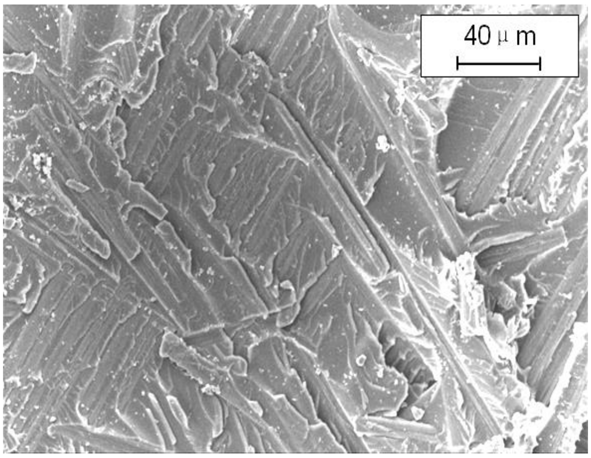


Fig.8 The fracture surface morphology of the laminates interleaved with NWCT

4 CONCLUSIONS

To improve the electrical conductivity and interlaminar mechanical properties of CFRPs laminates, nonwoven carbon tissue composed of short carbon fibers was prepared and incorporated into laminates as interleaves. The chopped carbon fibers in the interleaves were proved to be able to form the 3D network structure and efficiently simultaneously enhance the electrical conductivity, flexural mechanical properties and mode II interlaminar fracture toughness of CFRPs laminates compared with unmodified specimen. The SEM observation reveal that mode II interlaminar fracture toughness improvement can be explained by the fiber pullout and matrix deformation. The chopped fibers can be combined with pre-preging process to facilitate the end users to use the modified pre-pregs in a normal lay-up process. Thus, the fabrication technique of composites with chopped fibers reinforcement is validated in providing an applied prospect of the future research.

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