# ENHANCED IN-PLANE FRACTURE BEHAVIOUR OF NOTCHED CFRP REINFORCED WITH "KILOMETRE-LONG" CARBON NANOTUBE FIBRES

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## ABSTRACT

Although CNT fibres are theoretically known to have superior properties, research into their application in fibre-reinforced composites have largely been confined to providing additional interlaminar toughness. In this study, we introduced thin interlayers of continuous unidirectional CNT fibres for additional in-plane reinforcement and toughening of conventional autoclave-cured carbon fibre composites. Double notched specimens with and without CNT fibre reinforcing layers were tested to failure in tension. The results show that the introduction of relatively thin interlayers of continuous CNT fibres at local regions close to the notch tip increased the failure load and stress by 9%, and the failure strain by 15%.

## 1 INTRODUCTION

Although it is known for quite some time that individual carbon nanotubes (CNTs) exhibit desirable mechanical properties [1], much published research has been directed towards improving the stiffness and strength of pure resins or epoxies with discrete CNTs [2]. Some research has demonstrated the ability to grow CNTs onto conventional carbon fibres [3] or employ vertical CNT arrays as interlaminar bridging agents in CFRP laminates [4]. The vertically aligned CNTs have been successfully applied in hybrid CFRP sandwich composites to provide interlaminar reinforcement and enhance fracture toughness [5, 6]. Recently, an approach of hybridization of CFRP composites with thin interlayers of continuous carbon nanotube buckypapers or films has emerged to improve interlaminar properties [7-10], although the observed improvement in mechanical properties is not always consistent.

The strategy of bundling CNTs into fibres instead of films or buckypapers appears to be a preferred way to introduce the superior axial properties of CNTs into macroscopic structures. Recent advances in direct floating catalyst chemical vapour deposition (FC-CVD) synthesis [11] have enabled continuous and strong "kilometre-long" CNT fibres to be fabricated at high spinning rates in potentially sufficient quantities for structural applications. These CNT fibres exhibit not only desirable mechanical strength and stiffness, but also superior electrical [12] and thermal conductivities [13, 14], low density, excellent knot-efficiency [15], and high toughening potential [16]. Recently, CNT fibres with the intrinsic specific strength up to 2.0 GPa and stiffness up to 80 GPa have been synthesized [11]. With further treatment such as mechanical densification and/or polymer infiltration, tensile strength up to 1.5-3.0 GPa [17] and 3.6 GPa [18] has been reported. Therefore, continuous CNT fibres may emerge as candidates for hybridization with conventional synthetic or carbon fibre reinforced composites.

In this study, we introduce thin interlayers of unidirectional CNT fibres into a CFRP composite laminate made of commercially available prepregs in order to evaluate their influence on the in-plain properties and fracture behaviour of the hybrid composites.

## 2 EXPERIMENT

### 2.1 Unidirectional CNT fibre array manufacturing

The 'control' CF- and hybrid composites have been made from the commercially available thin T700/2510 unidirectional carbon/epoxy prepregs (CU-075, CGT International, Singapore) with a nominal thickness of 70 micron. CNT fibres were synthesized from methane as a carbon source via the floating catalyst chemical vapour deposition (FC CVD) process with details published elsewhere [18]. These CNT fibres had a linear density of ~0.3 tex and tensile strength and stiffness of not larger than 0.2 GPa and 13 GPa, correspondingly, and were used "as-synthesized" (without any additional densification or post-treatment). For each hybrid specimen, two unidirectional arrays of CNT fibers were made by manual winding 80-100 metres of CNT fibers on a metal mandrel. Care was taken to ensure that there were no fibre breaks, heavily-entangled fibre clusters, macro-inclusions or defects throughout the length of an array (Figure 1 a-c). After cutting from the mandrel, each unidirectional CNT fibre array of 1 cm-width and 7 cm-length consisted of well aligned and uniformly spread fibers (Figure 1 d,e).

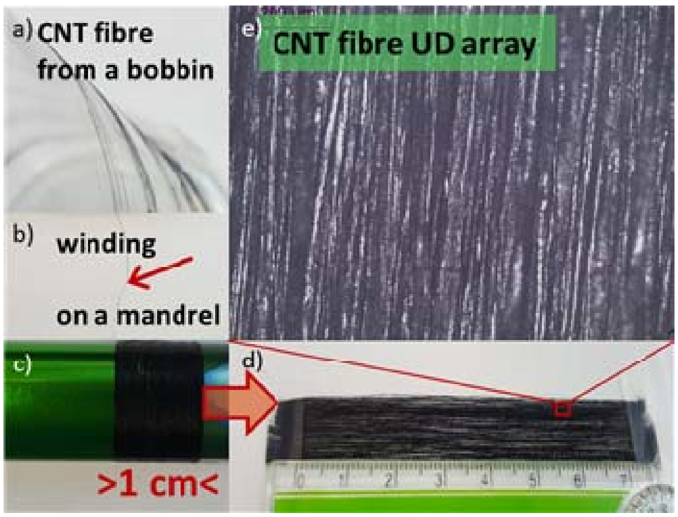


Figure 1: (a-c) Manufacturing of the CNT fibre array by taking a CNT fibre from its initial bobbin and winding it on a mandrel of 9 cm diameter until a width reaches 1 cm. d) 'Ready-to-use' CNT fibre array fixed on a siliconized paper for handling; and e) enlarged view showing good alignment of CNT fibres in the unidirectional array.

### 2.2 Composite processing

For each hybrid composite, two CNT fibre unidirectional arrays were positioned as the symmetrical interlayers according to the [45/90/-45/CNT layer/0/CNT layer/-45/90/45] layup. The control CFRP composite plates were prepared with the same layup without CNT layer inclusion. All the specimens were cured in the autoclave following the established curing procedure and water jet cut. The CNT interlayers were placed near the tips of the double notch where the stress concentration is greatest.

Figure 2: Manufacturing of the hybrid composites, including a) hand lay-up of the carbon fibre prepreg and the CNT fibre interlayers according to the [45/90/-45/CNT layer/0/CNT layer/-45/90/45] sequence; b,c) curing in the autoclave; d) cured hybrid composite plate, the yellow dashed rectangle illustrates location of the CNT layers; e) the water jet-cut notched specimen.

### 2.3 Mechanical testing and fracture analysis

A series of five control (without CNT reinforcement) carbon fibre thin ply composites and three hybrid composites with CNT fibre interlayers have been prepared for mechanical evaluation. Tensile testing was conducted by an Instron 8801 universal testing machine at a displacement rate of 1 mm/min. Strain and displacement on the specimen were measured using VIC2D digital image correlation (DIC) software based on the recorded displacement of random spackled pattern that were created on the surface of the specimen using spray paint.

The overall fracture surface developed in the control and hybrid composites has been analysed with the Leica optical microscope; the areas with CNT array localization have been also examined with the Helios Dual Beam / Focused Ion Beam microscope.

## 3 RESULTS AND DISCUSSION

Each unidirectional CNT fibre array contained several hundreds of individual CNT fibres mutually aligned and quite uniformly spread over the area of 1 by 8 cm (width and length, correspondingly), as shown in Figure 1. Total weight of an array measured with a high precision Sartorius ultra microbalance was about 0.016 g, resulting in the areal density of 20 g/m$^2$, which is significantly lower than that of a typical carbon fibre unidirectional prepreg (120 g/m$^2$). This low density is mainly due to the porous structure of the yarn-like CNT fibres [15], as they are formed by numerous CNT bundles intrinsically aligned in the axial direction. An excess of epoxy resin squeezed out from the carbon fibre prepreg plies during the autoclave composite processing was enough to infiltrate the CNT fibre layers, therefore no additional pre-infiltration was necessary. The thickness of the control (without CNT reinforcing layers) carbon fibre composites was 500 um consistently for a whole series. Inclusion of two CNT fibre interlayers gave an increase in the overall hybrid composite thickness of only ~35 µm (around 7%), which is half the thickness of a single CF ply (~70 µm). Accordingly, the thickness of each CNT fibre layer incorporated within the composite (being infiltrated with a resin and cured) was as small as ~15-17 µm.

The tensile tests of all the specimens were performed and the DIC was used to measure the displacement and verify that the maximum stress concentration originated in the area with CNT localization.

Figure 3 shows the load-strain and stress-strain curves for the control and hybrid composite specimens. Stress values were calculated by dividing the load by the composite nominal cross-sectional area at the double-notched area.

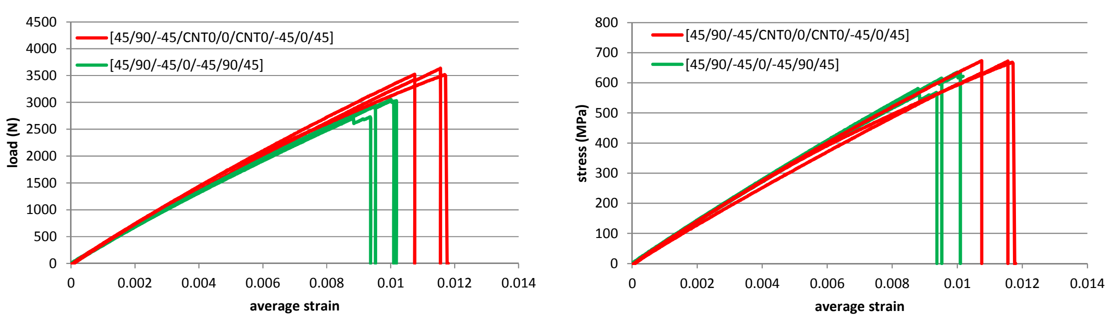


Figure 3: a) Load-strain and b) stress-strain curves for the control (green colour) and hybrid (red colour) composite specimens

The similar slope of the stress-strain curve indicates that the longitudinal modulus of the control composites has not been affected by inclusion of CNT fibre interlayers. The minimal effect on the longitudinal stiffness could be associated mainly with the high flexibility of the CNT fibres, which have a modulus of only up to 13 GPa.

However, it was observed that the relatively small (estimated to be less than 3% vol fraction) amount of continuous CNT fibres placed in a zero-direction increased the failure load from 2935 N to 3600 N when comparedto the control samples. This increases the average tensile strength by 9 %, from 613 MPa to 671 MPa, consistently observed for all the hybrid specimens. The initial CNT fibres possessed some deviation in the tensile properties, with the tensile strength and stiffness varied from 0.07 to 0.2 GPa and from 5 to 13 GPa, correspondingly. However, the nearly identical stress-strain curves, their slope, and resultant in-plane tensile strength of the hybrid composite indicate that the overall enhancement of the in-plane mechanical behaviour is governed only partially by mechanical properties of pristine CNT fibres (as-synthesized, before placing in a composite or resin infiltration). Factors such as polymer infiltration, CNT fibre compaction during the composite processing, CNT fibre/matrix bonding, as well as more consistent CNT fibre properties etc., could make significant contributions towards the hybrid composite reinforcement properties.

There was notable increase in the strain to failure of about 15%, suggesting the toughening potential of CNT fibre interlayers.

The post-failure fracture surfaces of the control and hybrid specimens have a similar pattern, with the main crack originated at the notched area (Figure 4a). Scanning electron microscopy analysis revealed not only the usual distinctive failures such as delamination, matrix failure, fibre breaks, fibre pull-outs, etc., one would expect in typical conventional CFRPs, but also multiscale delaminations and tensile failure within the interlayer of unidirectional CNT fibres (Figure 4b). Extensive pull out and re-arrangement of CNT bundles are also observed (Figure 4c).

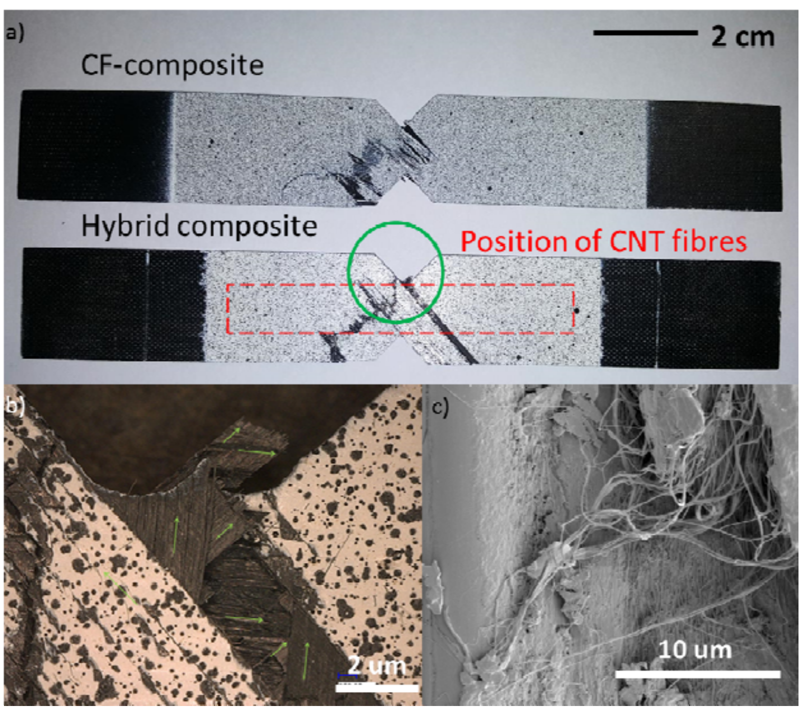


Figure 4: a) Double notched CF- and hybrid specimens after tensile test, location of the CNT layers is highlighted by the red dashed rectangle. b) Fracture surface of the hybrid composite at the notched area (enlarged from the green circle area above); c) fracture surface magnified with the FIB SEM microscope, showing CF and CNT fibres infiltrated with epoxy matrix and numerous CNT bundles

pulled-out in the tensile test.

## CONCLUSION

In this study, we demonstrate in-plane strengthening and toughening of carbon fibre reinforced composites by the introduction of highly aligned and continuous unidirectional CNT fibre thin interlayers. Tensile tests of the double notched composite specimens demonstrate an almost 9% increase in failure stress without decrease of stiffness. The increase in the strain to failure of about 15% shows the toughening potential of CNT fibre interlayers for hybrid composites.

Future further enhancement and optimization of the FC-CVD synthesis process are anticipated to produce even stronger and stiffer CNT fibres with better consistency in quality. This could have more pronounced improvements on the mechanical properties of future hybridized and lightweight composite structures.

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